

## Chapter 7

# Coal-Fuelled Combined Cycle Power Plants

Combined cycle power plant, when used as a generic term, refers to a plant which converts heat into mechanical energy in a combined gas and steam turbine process. Combined cycle processes with coal gasification or coal combustion turn solid fuels into a fuel gas or a hot pressurised gas which is then used in the gas and steam turbine processes. Coal-fuelled combined cycle plants will be discussed in detail in the following sections. A start will be made by describing the basic technical features and the characteristic data of combined cycle power plants fuelled by natural gas for the purposes of comparison to coal.

### 7.1 Natural Gas Fuelled Combined Cycle Processes

The combined cycle process offers a number of advantages over the simpler steam – water only process. These are

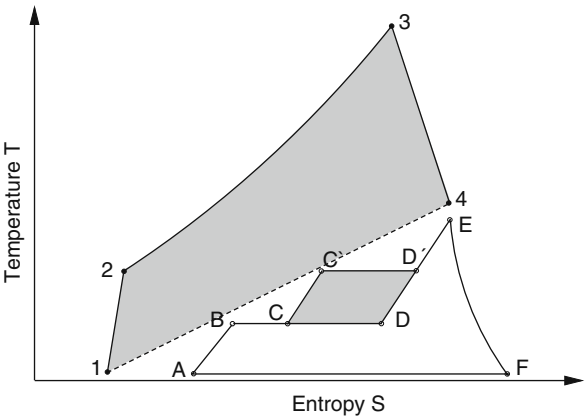
- highly efficient generation of electrical power,
- a straightforward process,
- low investment costs and
- a smaller environmental impact.

The only requirement is a fuel gas which is suitable for gas turbines, for example natural gas.

The high efficiency results from combining the high-temperature gas turbine process with the low-temperature steam process. The fuel is fed to the process only via the gas turbine combustion chamber. While it is being combusted with compressed air, hot flue gas is produced under pressure in the combustion chamber. The gas turbine then converts the energy from the pressurised hot flue gas into mechanical energy. This causes the gas to expand, having lost most of its pressure, at low temperature at the turbine outlet. The residual heat from the flue gas has a temperature of 500–600°C and is transferred to the downstream steam process.

In a natural gas fired combined cycle, about two thirds of the electrical power is produced in the gas turbine and one third in the steam turbine. The gas turbine

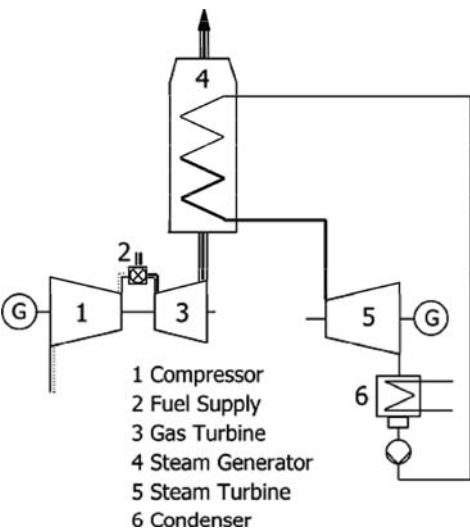
**Fig. 7.1** Combined cycle process in a  $T-S$  diagram with a gas turbine process (1-2-3-4) and a single pressure (A-B-C-D) or dual-pressure steam process (A-B-C'-D'-E-F)



process is normally called the topping process and the steam process the bottom process. A  $T-S$  diagram of the two cycle processes is given in Fig. 7.1.

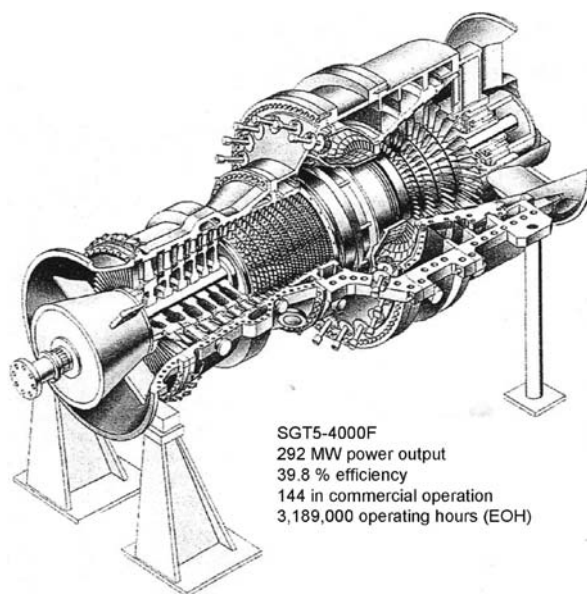
Figure 7.2 shows a natural gas fuelled combined cycle process. The gas turbine installation consists of a compressor, a combustion chamber and a gas turbine. The steam plant consists of a heat recovery steam generator (HRSG), a steam turbine and the subsequent steam – water cycle.

Gas turbines are available commercially as standard products consisting of an integrated gas turbine, compressor and combustor. They feature a high power density and fixed design data, to which only minor changes can be made. Figure 7.3 shows a sectional view of a modern gas turbine with a capacity of 292 MW<sub>el</sub>. The largest gas turbines today have a capacity of around 340 MW<sub>el</sub> (Schöler 2007; Ratcliff et al. 2007).



**Fig. 7.2** Diagram of the combined cycle process

**Fig. 7.3** State-of-the-art gas turbine (Source: Siemens)



Natural gas has an adiabatic combustion temperature of about  $2,200^{\circ}\text{C}$ . For this reason, the furnace of the gas turbine is operated at a high excess air level in order to reduce the flue gas temperature to the permissible inlet temperature of the gas turbine itself. The flue gases in the newest gas turbines enter the turbine at a temperature of around  $1,400\text{--}1,500^{\circ}\text{C}$ . The turbine blades closest to the entry point, in particular, are therefore subjected to high thermal and mechanical stresses, as well as chemical stress from oxidation and corrosion.

Since the metallic turbine materials currently in use can only be exposed to surface temperatures of up to around  $950^{\circ}\text{C}$ , the first stages of a gas turbine are cooled by air from the compressor. Over the last decades cooling has evolved from convection over impingement to film cooling. The blades are made from a composite material; the base material provides the mechanical properties, and one or more surface-protective coatings ensure corrosion and oxidation resistance. Thermal barrier coatings can also offer thermal insulation and allow higher gas turbine inlet temperatures (Bohn 2007).

There is no standard definition of the turbine inlet temperature (TIT). The term can, in fact, refer to different temperatures. The three definitions of relevance are as follows:

- (1) The temperature at the outlet from the combustor (i.e. at the inlet to the first turbine stator).
- (2) The temperature at the outlet from the first turbine stator (i.e. at the inlet to the first rotor). At this point, the temperature is typically  $40\text{--}70^{\circ}\text{C}$  lower than in definition 1 due to the cooling air or steam for the first stator. This definition of the TIT is used by General Electric.

- (3) The calculated temperature (not physical) that would result from mixing all the cooling air with the combustor exhaust gas. This temperature is typically 70–110°C lower than definition 2. Siemens uses this definition, which is also known as the “ISO TIT”.

It should be added that all three definitions would be identical for a gas turbine without cooling (Maurstad 2005). In this book, definition 1 (TIT) and definition 3 (ISO TIT) will be used.

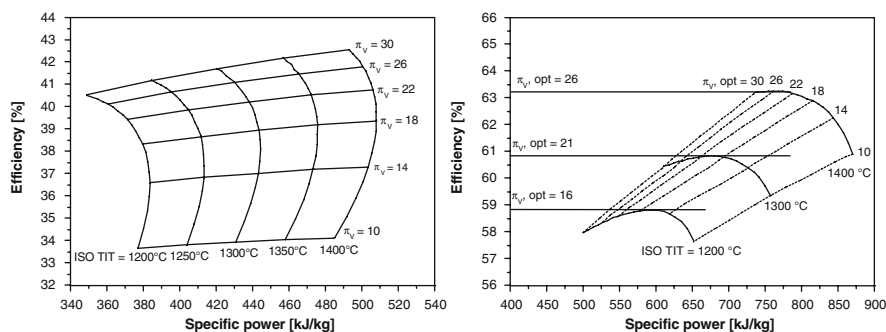
Given a TIT of roughly 1,400°C at the outlet from the combustor, the ISO temperature calculated for a modern gas turbine is around 1,200°C. The highest ISO gas turbine inlet temperature is around 1,320°C in current practice.

Figure 7.4 shows how the efficiency is influenced by the design parameters (e.g. the ISO turbine inlet temperature and pressure ratio of the gas turbine), both for the gas turbine itself and for the turbine in the gas/steam combination of a combined cycle power plant.

When the gas turbine is operated by itself, the turbine’s pressure influences the efficiency, while the inlet temperature influences the output. With a constant gas turbine inlet temperature, the efficiency rises as the pressure increases. This is because the flue gas outlet temperature behind the turbine and hence the losses decrease. If the pressure rises above the optimum level, the energy required for compression increases more quickly than the additional gain in the gas turbine’s power output.

The gas turbine inlet temperature has only a minor impact when the gas turbine is being operated alone. This is because the higher temperature of the heat input is partly compensated for by the higher temperature of heat dissipation. In combined cycle operation, the inlet temperature of the gas turbine has a discernible effect on efficiency. The gain in efficiency obtained by the higher temperature of the heat input is conserved. This is because the waste heat of the high flue gas temperatures is used in the heat recovery process.

The optimum pressure of a gas turbine for the combined cycle process is lower than for a gas turbine operated by itself, because the steam process only achieves a high efficiency at flue gas temperatures above 600–650°C, which generates

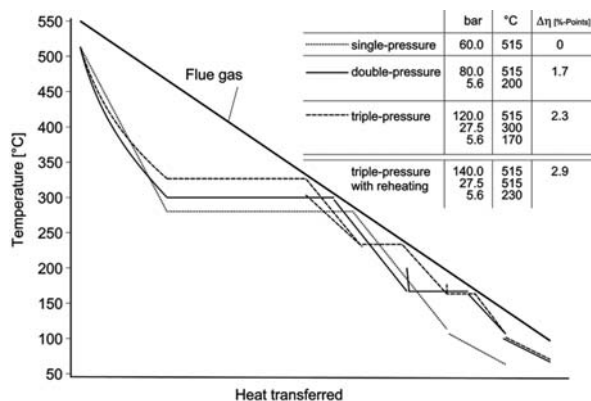


**Fig. 7.4** Impact of pressure and the gas turbine inlet temperature (ISO) on the efficiency and output of a gas turbine and a combined cycle process (Kloster 1999)

state-of-the-art high live steam temperatures of 550–600°C. As a result, a gas turbine designed to be operated by itself and which features a high efficiency, a high pressure and a low gas turbine outlet temperature does not necessarily result in a high efficiency when used in a combined cycle. Flue gas temperatures below 600°C lead to a decrease in the efficiency of the steam turbine process higher than the efficiency increase in the gas turbine. On the other hand, flue gas temperatures higher than the level required for optimum efficiency lead to exergy losses in the heat recovery process, because the steam temperature is limited by the materials. The outlet temperatures of modern gas turbines, however, are close to those temperatures which make high-output steam production possible (Kail and Rukes 1995; Kloster 1999).

The number of gas turbine types available from manufacturers is limited. This places restrictions on how combined cycle power plants can be designed. Unlike steam turbines (which are designed for a given steam cycle on a case-by-case basis), gas turbines on the market feature specific capacities only. Modifying the gas turbine is expensive, because significant design work and manufacturing modifications are involved, in particular for the compressor.

Once a gas turbine has been selected, the main design parameters for the downstream heat recovery process (the gas mass flow and the gas turbine outlet temperature) are specified. A high total plant efficiency is produced when the heat is transferred from the gas turbine flue gas to the steam – water cycle with minimum energy losses. In practice, this can be achieved by generating the steam at various pressures, so that the difference in temperature between the flue gas to be cooled and the heat-receiving medium (and hence the energy losses) can be decreased. This is shown in Fig. 7.5 (Riedle et al. 1990). Today, large plants optimised for high electrical efficiency use a triple-pressure heat recovery process. In addition to the number of pressure stages, the heating surfaces of the heat recovery steam generator (HRSG) allow the energy losses of the waste heat transfer to be diminished. Selecting the number of pressure stages and the design of the heat exchanger are subject to cost-effectiveness considerations: the additional costs are pitted against the savings resulting from the higher efficiency (Warner and Nielsen 1993).



**Fig. 7.5** Temperature course in a waste heat boiler (Riedle et al. 1990)

**Table 7.1** Possible development of combined cycle processes (Bohn 2005)

Parameter	Reference	Phase 1	Phase 2	Phase 3
Efficiency	57.4%	61.7%	63.3%	65.0%
Combustion exit temperature	1,500°C	1,500°C	1,520°C	1,520°C
Cooling GT	21.46%	11.7%	12.4%	9.9%
ISO turbine inlet temperature	1,172°C	1,432°C	1,439°C	1,473°C
Max. substrate temp. (GT)	850°C	900°C	950°C	990°C
Max. material temp. (ST)	560°C	580°C	595°C	650°C
Required cooling steam	–	2.2%	1.0%	0.3%

State-of-the-art gas turbine technology, designed for gas turbine only plants, achieves efficiencies of up to 38%. In a combined cycle plant with an optimised heat recovery process, efficiencies rise to 58% (Jopp 2005). A new gas turbine is currently being tested with an efficiency of around 40% as a stand-alone gas turbine and above 60% as part of a combined cycle plant (Schöler 2007). Gas turbine technology has undergone continuous, high-level development in recent years. A further rise in the ISO gas turbine inlet temperature, and hence in the gas turbine efficiency, is expected in the future. A higher ISO TIT results in higher gas turbine outlet temperatures and promotes the use of advanced live steam conditions. Table 7.1 shows potential developments in the coming years (Bohn 2005; Bohn 2007).

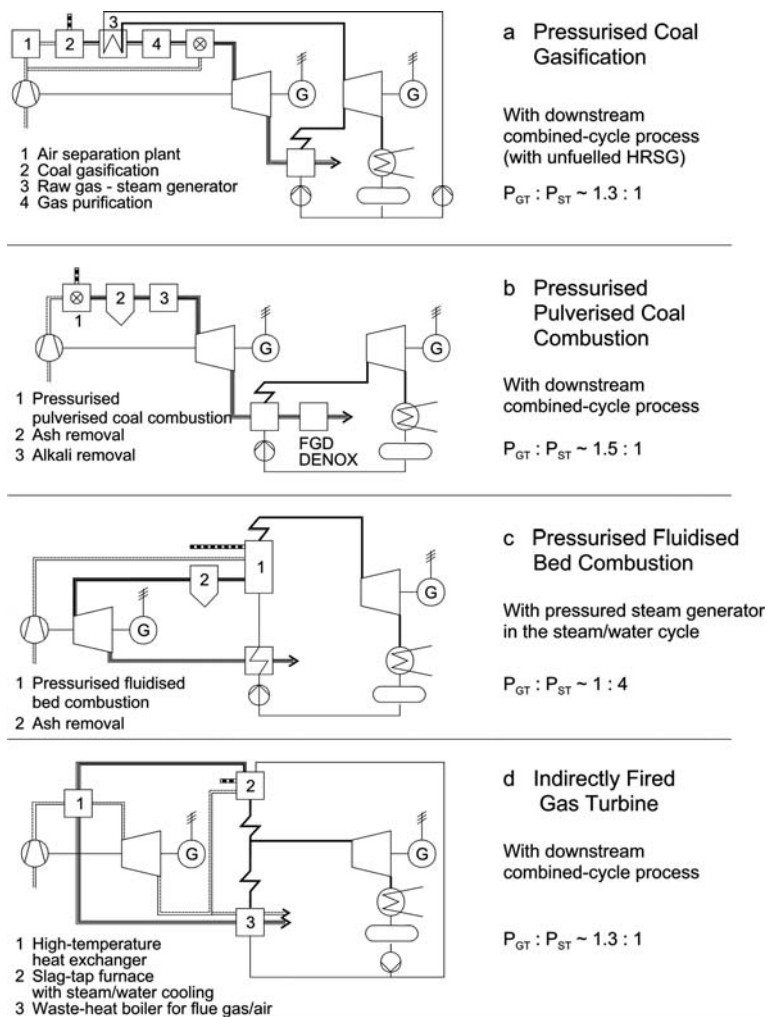
The high efficiencies of the combined cycles shown above are only possible with a clean fuel such as natural gas. Otherwise, the purity requirements of the gas turbine for the working medium (pressurised hot flue gas) cannot be met.

## 7.2 Overview of Combined Processes with Coal Combustion

### 7.2.1 Introduction

For coal-based combined cycle processes, the solid fuels must first be converted into a fuel or a hot gas suitable for gas turbines. Impurities in the fuel prevent it from being used directly in today's gas turbines. Compared to the natural gas fired combined cycle process, more steps are required (COORETEC 2003; JBDT 1992; Rukes 1993; Wittchow and Müller 1993; Jahraus and Dieckmann 1989; Böhm 1994). The processes shown in Fig. 7.6 have been investigated, tested and partially implemented on an industrial scale in order to put a coal-based combined cycle into practice. They are

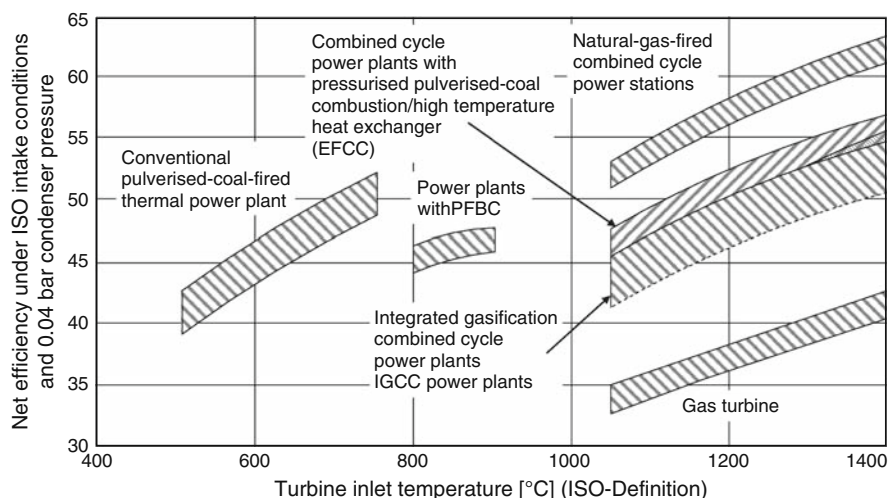
- integrated gasification combined cycle (IGCC),
- combined cycle with pressurised fluidised bed combustion (PFBC),
- combined cycle with pressurised pulverised coal combustion (PPCC) and
- externally fired combined cycle (EFCC).



**Fig. 7.6** Coal-based combined cycle processes (Böhm 1994)

In the integrated gasification combined cycle, the process starts by transforming the solid fuel into a fuel gas at high temperatures in a gasifier. The gas then has to be purified. Following this, the fuel gas is burned in the gas turbine. The fuel gas produced is cooled by water/steam for the flue gas cleaning step. This results in a low ratio (about 1.3:1) of the gas to the steam turbine power outputs, and hence to a lower efficiency compared to the natural gas fired combined cycle.

In the combustion-based combined cycles of pressurised fluidised bed combustion (PFBC) and pressurised pulverised coal combustion (PPCC), the fuel is completely combusted under pressure. Before entering the gas turbine, the hot flue gas has to be cleaned so that it meets the required gas purity standard. Whereas in



**Fig. 7.7** Efficiency of combined cycle processes depending on the gas turbine inlet temperature

the PPCC process a 1.5:1 ratio of the output of the gas and steam turbines can be achieved, the PFBC process, with its steam – water cooling, yields a mere 1:5 ratio of the gas and steam turbine outputs. The ratio of the gas to the steam turbine power outputs correlates directly with the efficiency of the overall cycle; the higher the ratio, the higher the efficiency.

There is an essential difference between the combined cycles with gasification and with combustion: the volumetric flow to be cleaned is 10 times higher for combustion. In a power plant with an integrated gasification combined cycle, the fuel gas

**Table 7.2** Comparison of power plant processes

	Steam power plant	Natural gas fired combined cycle	Coal gasification combined cycle	Pressurised fluidised bed combustion	Pressurised pulverised coal combustion	Externally fired combined cycle
Stage of development	State of the art	State of the art	Can be demonstrated	Being launched on the market	In development	In development
Relative investment costs	100%	30%	110–120%	100%	?	?
Relative availability	> 90%	> 90%	Lower than steam power plant	> 90%	?	?
Efficiency (1,250° C ISO)	46%	58%	51%	45% (51%) <sup>a</sup>	53	53

<sup>a</sup> Hybrid process



flow can be cleaned at a lower temperature than that of gasification. By contrast, in the combustion-based combined cycles with pressurised fluidised bed combustion and pressurised pulverised coal combustion, the flue gas has to be cleaned at temperatures above the gas turbine inlet temperature. Otherwise, there would be a high loss in efficiency.

In the externally fired combined cycle (EFCC) process, a high-temperature heat exchanger is used to avoid the problems of hot gas cleaning. The energy yielded by combustion is transferred to a clean working medium in a heat exchanger. This working medium, which is suitable for use in a gas turbine, charges the turbine.

These different methods are at differing stages of development. While both PFBC and IGCC are already being used in industry, the EFCC and PPCC processes have not been implemented at industrial scale.

In Fig. 7.7, the efficiencies of coal and natural gas based combined cycles are compared with each other and with the conventional steam power plant at a specific turbine inlet temperature. Table 7.2 shows a comparative evaluation.

### ***7.2.2 Hot Gas Purity Requirements***

The purity of the working medium for the gas turbine required by gas turbine manufacturers is a parameter which determines how the conversion process is designed and which purification steps are selected. When evaluating impurities in the fuel gas or hot flue gas, a distinction must be made between components that lead to

- high-temperature corrosion, erosion and deposits in the gas turbine,
- corrosion at the cold end of the heat recovery steam generator or
- undesirable emissions.

The damage to the flue gas charged gas turbine caused by corrosion, erosion and deposits shall be discussed below, because monitoring these problems is decisive in ensuring the success of coal-fired combined cycles.

Deposits on the turbine blades lead to a decline in the turbine efficiency. They can be removed by scrubbing the gas turbine. Such a process uses water jets installed in the combustion chamber. Desalted water is sprayed into the chamber under pressure and reaches the turbine via the air flow. This water washes away the water-soluble compounds and penetrates the pores and crevices of water-insoluble compounds. When the gas turbine heats up, the water in the pores and crevices evaporates. The resulting steam pressure causes the deposits to spall (JBTD 1992).

Erosion and corrosion wear the turbine down and lead to a reduction in the turbine's lifetime and efficiency. Solid as well as liquid particles reach the turbine blades in a number of ways (Thambimuthu 1993):

– Inertial impaction:

Large particles do not follow the gas flow due to their inertia. The particles which hit the blades cause erosion but can also add to deposits. Impinging particles can

also carry deposits away again under certain conditions. Particles smaller than  $5\text{ }\mu\text{m}$  or so follow the gas flow and therefore do not cause erosion.

- Turbulent and Brown's diffusions:

Inertia is not the only factor which can cause particles to deviate from their course. Turbulent and Brown's diffusions can have a similar effect. In the case of turbulent diffusion, the particles are caught by the eddies of the turbulent boundary layer flow. In Brown's diffusion, the kinetic energy of the gas molecules is transferred to small particles. Both turbulent and Brown's diffusion phenomena catch even the smallest particles, which are deposited over the whole inner surface of the gas turbine.

- Thermophoresis:

Thermophoresis refers to transportation by thermal diffusion as a result of thermal gradients between the gas and the surfaces. While thermophoresis can be ignored in the case of low gas turbine inlet temperatures, it can play a significant role where blade cooling (required for higher tube inlet temperatures) is involved.

Of particular importance with regard to contamination and corrosion are sodium and potassium. These gaseous alkalis are released during combustion, and even small traces of them can shorten the lifetime of the gas turbines. Gaseous alkalis condense in the turbine while it is cooling down. They then form molten alkali sulphates (in compounds using  $\text{SO}_2$  from the flue gas, for example) which are deposited on the turbine blades or on ash particles. Alkalis cause the ash fusion temperatures to fall, and this in turn causes some or all of the ash particles to melt, either before or after they reach the blade. In the latter case, they form deposits on the blades. Melting or sintering of the ash causes deposits to form on the blades that are difficult to remove (Thambimuthu 1993).

The molten alkali compounds attack the gas turbine blades severely. By means of high-temperature corrosion, they destroy the oxidic protective layers of the parts they come in contact with. This causes an intense corrosive attack on the unprotected base material, resulting in a drastic shortening of the lifetime of the blades. The rate of corrosion depends on the chemical composition of the deposits; the greater the alkali content, the faster the rate of corrosion. The available information on the corrosive impact of alkalis is based on experience of the combustion of oil distillation residues and heavy fuel oil in gas turbines. Literature on this issue often uses the limit of  $0.024\text{ mg/kg fuel}$ , set by General Electric, for the combustion of oil distillation residues, though it varies depending on the fuel used. Even if it seems impossible to apply this data directly to the conditions of coal-fuelled firing, they do form a basis for specifying the alkali requirements to be met by the hot gas.

Certain metals such as vanadium, lead and zinc can cause the same destruction of protective layers as do alkalis. Chlorine, fluorine and their acids can wear away the protective layers by forming gaseous chloride and fluorides (JBDT 1992). Alkaline earth metals can lead to hard deposits on the turbine blades (Hannes et al. 1989).

The limiting values for turbines depend on the gas turbine inlet temperature. Lower permissible limits are reported for higher temperatures. Much less stringent requirements are expected after PFB firing due to the gas turbine inlet temperatures, which are lower in comparison to pressurised pulverised coal firing.

**Table 7.3** Permissible guideline concentrations for dusts and trace elements in the hot gas for gas turbine V94.3 (now SGT5-4000F) (data from Jansson 1996; Mitchell 1997)

Dust	Total	[mg/kg]	1
Dust distribution	> 10 $\mu\text{m}$	% by wt	0
	2–10 $\mu\text{m}$	% by wt	7.5
	0–2 $\mu\text{m}$	% by wt	92.5
Trace elements	Ca	[mg/kg]	0.4
	V + Pb	[mg/kg]	0.01
	Na + K	[mg/kg]	0.01

Table 7.3 gives the permissible flue gas concentrations in front of the turbine for the Siemens V94.3 (now SGT5-4000F) gas turbine at a gas turbine inlet temperature of 1,120°C (ISO) which are required after combustion of natural or coal gas. The table shows the limits for the total particulate matter content, the maximum size of particles and the concentrations of heavy metals – lead (Pb) and vanadium (V) – the alkaline earth metal calcium (Ca) and the alkali metals sodium (Na) and potassium (K) in the flue gas (Jansson 1996). For combined cycles with pressurised coal firing, limits comparable to natural gas fuelled gas turbines are used as a basis.

The fact that pollutants can also be sucked in together with the combustion air must be taken into consideration as well. At coastal locations, for instance, the marine salt contained in the fresh air can make a major contribution to the alkali load in the process.

Although gas turbines have been operated with hot gas generated by PFB combustion for several years now, no detailed data on erosion, deposits and corrosion, or relevant limits for prevention of such damage, is available. The values achieved with the hot gas filters currently used for PFB furnaces, i.e. 250–650 mg/Nm<sup>3</sup>, reveal much higher concentrations of particulates in the flue gas than the values given in Table 7.3. All particles are smaller than 10  $\mu\text{m}$  and the mean diameter is 2–3  $\mu\text{m}$  (Jansson 1995b). The concept developed by ABB (now Alstom) is based on a modified robust gas turbine, designed to minimise erosion.

The technical data currently available is provided by experimental plants with gas turbine cascades. In order to limit the erosion caused by hot gases in fluidised bed combustion (FBC) furnaces, limits are suggested for concentrations of particles larger than 4  $\mu\text{m}$  as such (Stringer 1989):

- > 20  $\mu\text{m}$ : 1 mg/kg
- 10–20  $\mu\text{m}$ : 1 mg/kg
- 4–10  $\mu\text{m}$ : 10 mg/kg

Particles smaller than 4  $\mu\text{m}$  are likely to appear but they will not cause erosion, contamination or corrosion. At lower gas turbine inlet temperatures in PFB furnaces, concentrations of particulates up to 100 mg/Nm<sup>3</sup> are considered tolerable as long as all particles are smaller than 5  $\mu\text{m}$  (Emsperger and Brückner 1986).

The alkali emissions from PFB furnaces can be one or more orders of magnitude higher than the 0.024 mg/kg fuel indicated as a limit for turbine corrosion for combustion of oil distillation residues. Although the installed PFB furnaces are not equipped with an alkali remover, the reported corrosion is mild.

**Table 7.4** Required flue gas purity for pressurised pulverised coal combustion

Dust content	[mg/Nm <sup>3</sup> ]	3
Maximum particle diameter	[μm]	< 3
Gaseous alkalis	[mg/Nm <sup>3</sup> ]	0.01

The values in Table 7.4 for the required purity of the flue gas at the turbine inlet temperature for the pressurised pulverised coal firing concept are somewhat higher than the values given in Table 7.3 (Hannes 1986; Förster et al. 2005).

### ***7.2.3 Overview of the Hot Gas Cleaning System for Coal Combustion Combined Cycles***

In a combined cycle with coal gasification, cold gas cleaning of the fuel gas is the standard procedure. By contrast, hot flue gas cleaning, an efficiency-improving option, is essential for combustion-based combined cycles. FBC systems can operate with no more than dust removal, whereas pressurised pulverised coal firing also requires alkali removal. The removal of pollutant gases (sulphur dioxide or nitrogen oxide, for example) which the gas turbine can tolerate can take place either before or after the gas turbine. While sulphur dioxide removal can take the form of an in situ process in fluidised bed firing, pressurised pulverised coal firing requires desulphurisation after the heat has been recovered in the steam generator.

The choice of the dust removal technique depends on the fusion behaviour of the fuel ash. Below the ash deformation temperature, fly ash is separated in solid form; at temperatures above ash fluid temperatures, it is separated as liquid slag. In the temperature range of roughly between 900 and 1,300°C, hot gas cleaning of ash is possible in theory. However, difficulties will arise when cleaning the filter components, because the particles stick to the equipment.

Table 7.5 offers an overview of the possible dust removal techniques (Weber and Pavone 1990; Weber et al. 1993; Pruschek et al. 1990). They can be classified as follows:

- Mass force separators
- Wet scrubbers
- Filter separators
- Electrostatic precipitators (ESPs)

The class of filters used for *separation by mass force* comprises all separators that use only mass forces to clean the gas, i.e. gravity, inertia or centrifugal force. The names of the various separators are therefore derived from the effective force in the case in question. The majority of mass force separators are straightforward in design and pose few engineering problems. Compared to other dust-collecting devices, they are cost-effective and easy to service and operate. However, they are not suitable for the removal of soft or sticky particles. They are well suited to the removal of liquid slag at temperatures above 1,500°C, though material-related problems may arise.

**Table 7.5** Summary of temperature windows for use of particulate matter collection technologies

Flue gas temperatures	Mass force separator	Wet scrubber	Filter separators	ESP
< 900°C (solid)	Possible	Possible with suitable scrubbing liquids	Possible with ceramic filter elements	Possible
850–1,300°C (melting range)	Ash removal not possible because particles are sticky	No suitable scrubbing agent known	Not possible because particles are sticky	Not possible because particles are sticky
Above 1,300°C (liquid)	Possible; material problems	Possible; material problems	Not applicable for liquids	Not possible

*Wet separators* can, in principle, be used for high-temperature cleaning if a suitable, thermally stable scrubbing liquid is used at low vapour pressures. The wet scrubbing process is complicated by the treatment of the scrubbing liquid and the material used. At temperatures up to 850°C (and possibly higher), this method is more complicated and expensive than other dust-collecting methods. The scrubbing medium used for the pressurised pulverised coal fired furnace is liquid slag. When injected into a venturi scrubber, it helps the liquid slag droplets to agglomerate, thus enhancing the removal efficiency in a downstream cyclone. For more information, see Sect. 7.4.2.

In their various technical designs, *filter separators* generally form suitable high-temperature particle collectors if suitable thermally stable filter materials are available and the particles to be separated are solid. If the particles are sticky, removing the particles from the filter material becomes a problem. For temperatures above 1,250°C, the principle is not applicable because of the liquid consistency of the ash. In contrast to the other particulate collection technologies, not only do the electrostatic precipitators in the high-temperature range present material and construction problems, but the physical conditions for collection must also remain stable at extreme gas temperatures. Investigations at temperatures up to 1,000°C show that the production of charge carriers (required for particle removal) is possible. At temperatures of 1,300°C or above, the ESP principle cannot be applied due to the conductivity of the flue gas.

The individual methods for removing solid particles are discussed in Sect. 7.3.2 in relation to pressurised fluidised beds; the removal of molten ash particles is discussed in Sect. 7.4.2 in relation to pressurised pulverised coal firing.

### 7.2.4 Effect of Pressure on Combustion

Combustion of the residual char is a major factor in determining how long coal combustion will take. This is described in detail in Sect. 5.2. The effect of pressure on the burning speed of the char depends on whether chemical reactions or transport

processes during combustion of the solid char influence the reaction velocity. At low temperatures, chemical reactions slow down the reaction velocity. Given that the chemical reaction velocity is proportional to the oxygen concentration, an increase in pressure will accelerate char combustion. In contrast, pressure exerts only a minor influence on the diffusion-controlled reaction.

A clear improvement of the combustion is therefore expected in the temperature range of the fluidised bed combustor. With its higher temperatures, however, the conditions under which pulverised fuel combustion occurs leave little room for a substantial acceleration of combustion. In this case pressure cannot raise the speed of combustion, but it does influence the dimensions of the furnace. Figure 7.8 shows the effect of pressure, as calculated for different temperatures (Gockel 1994).

If the furnace has the same geometry as in the process at ambient pressure, the air mass flow (which enters the furnace at constant inlet speeds) will increase proportionally to the pressure. The fuel mass flow will then change accordingly if the carrier gas has a constant fuel load. Since the pressure does not cause any change in speed, the residence time required for the purpose of combustion remains constant. From a combustion perspective, the output can be increased proportionally to the pressure as long as the furnace volume remains the same. Alternatively, the volume can be decreased proportionally to the pressure with no change in output and without taking the pressure effect on the speed of combustion into account.

If the furnace has to dissipate heat in addition to releasing it (as in the case of stationary fluidised bed), the impact of pressure on the heat transfer also plays a role in determining the furnace's dimensions.

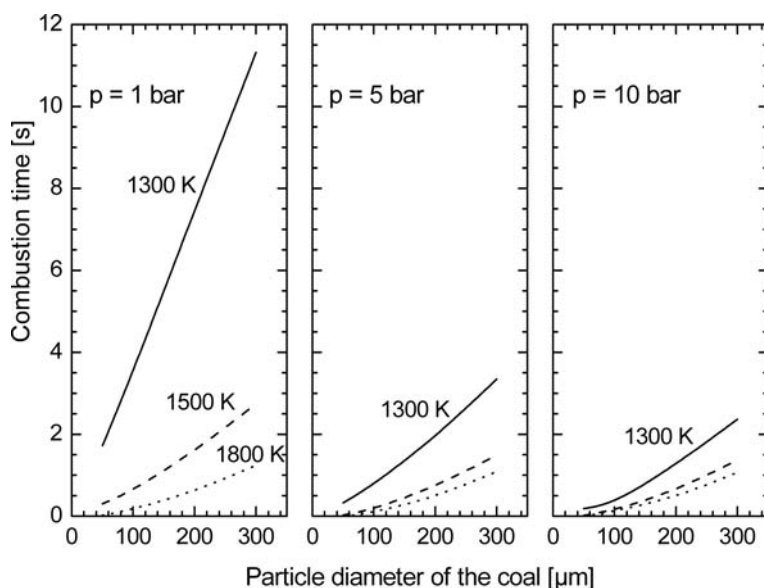


Fig. 7.8 Effect of pressure on combustion (Gockel 1994)

## 7.3 Pressurised Fluidised Bed Combustion (PFBC)

### 7.3.1 Overview

The pressurised fluidised bed is one method of using coal as a fuel in a combined cycle process. The solid fuel is burned in a bubbling or circulating fluidised combustion bed at temperatures between 850 and 950°C and pressures of up to 16 bar. Following cleaning, the hot flue gas is brought to the gas turbine.

The optimum temperature in the fluidised bed is determined by the requirement that it must remain below the ash deformation temperature at all times, in order to prevent agglomerations from forming in the fluidised bed and in order to achieve optimal desulphurisation. Therefore it is necessary to cool the fluidised bed, as the adiabatic combustion temperature of coal is above 2,000 °C. Various methods can be used for the purpose of cooling: operation at a high excess air level, air cooling or vapour – water cooling (Emsperger and Brückner 1986). In natural gas fired combined cycles, by comparison, the gas turbine inlet temperature (which is much higher than the fluidised bed though) is set by operating at 100% excess air. Figure 7.9 shows the different cooling process variants, which are described below:

- *Adiabatic pressurised fluidised bed:*

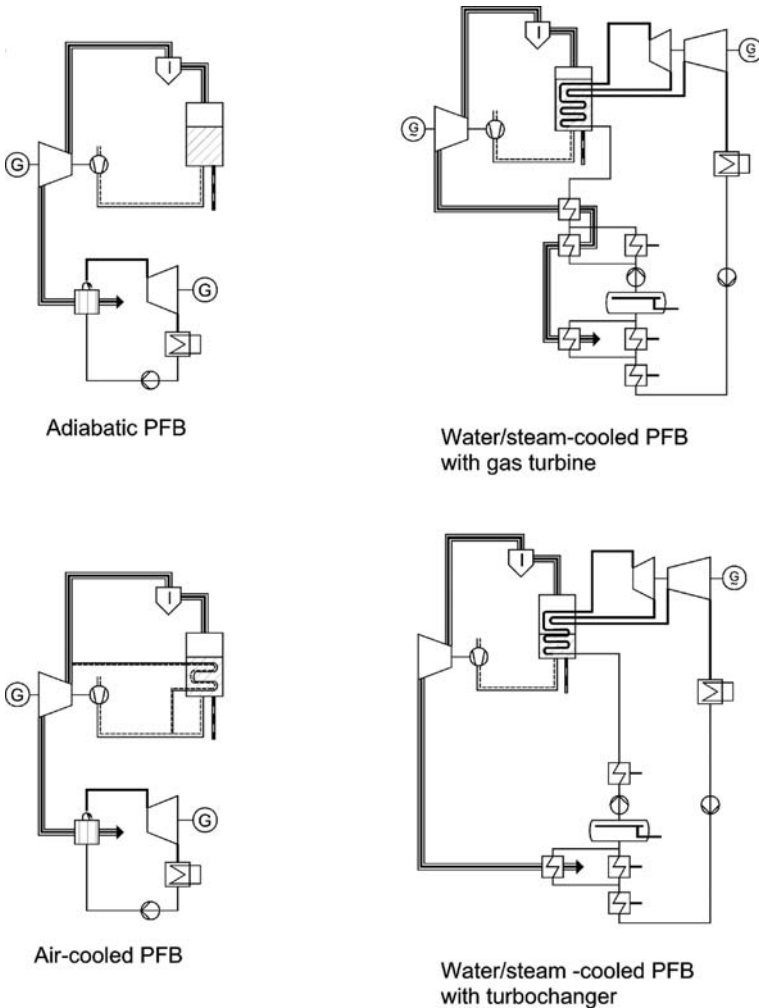
An adiabatic fluidised bed with no in-bed heating surface needs an excess air level of 300% (an air ratio of 4) in order to limit the fluidised bed temperatures. This results in a furnace with large dimensions and particulate removal downstream. The ratio of the gas turbine output to the steam turbine output is about 2:1. The adiabatic fluidised bed has been studied conceptually, but as yet a plant has been neither planned nor constructed.

- *Pressurised fluidised bed with an air-cooled heating surface:*

In this configuration, compressed air is heated to about 700°C in the in-bed heat exchanger and mixed with the cleaned flue gas before the gas turbine. Due to the lower air temperature, the gas turbine inlet temperature drops below the temperature of the fluidised bed, resulting in a lower efficiency. The ratio of the gas turbine output to the steam turbine output is about 2:1. This variant presents some major drawbacks, such as the poorer heat transfer of air as a cooling medium (compared to steam and water) and the higher material temperatures.

- *Pressurised fluidised bed with steam – water cooling:*

Steam – water cooling is the method used in today's PFBC furnaces due to the more compact, and hence more cost-effective, design of these furnaces. Therefore, only this variant shall be discussed in more detail. A disadvantage is the fact that the gas turbine makes no more than a minor contribution (one fifth to one quarter) to the total power output. The efficiency depends on the steam conditions used. In the case of a high-quality steam process, the efficiency is higher than that of the adiabatic fluidised bed; in the case of low-quality steam conditions, the efficiency is lower.



**Fig. 7.9** Cooling of PFBC furnaces (Emsperger and Brückner 1986) and amendments

– *Pressurised fluidised bed with steam – water cooling and turbocharger:*

In this configuration, the flue gases are cooled down by convective heat exchange surfaces to about 400–500°C before particulates are collected. This avoids the problem of hot flue gas cleaning at temperatures of around 850°C. At these low gas turbine inlet temperatures, however, the gas turbine only provides the power necessary to drive the compressor. The advantage of this configuration is limited to its compact design. Its efficiency depends solely on the steam process (Emsperger and Brückner 1986; Dibelius and Pitt 1989). Such a plant, with a thermal capacity of 40 MW and a maximum pressure of 4 bar, was used between 1984 and 1989 in the thermal power station operated by the Aachen University of



Technology. Its purpose was to prepare design data and test components as part of a research project (Terhaag et al. 1995; Thelen 1993; Emsperger and Brückner 1986).

Figure 7.10 shows various flue gas cleaning flow diagrams for PFBC furnaces (Thambimuthu 1993). Flue gas cleaning works at temperatures between 800 and 900°C. Its purpose is to prevent fouling and to protect the gas turbine from erosion and corrosion. Commercial plants mainly use cyclone separators. Ceramic hot gas filters have been tested in the Tidd, Wakamatsu and Escatrón plants. However, this technology is not considered to be ready for commercial PFBC plants. Alkali removal is not used in commercial PFBC plants (McMullan 2004; Sasatu et al. 2001; Wright et al. 2003; Wu 2006).

The upper limit of FBC temperatures in order to avoid bed agglomeration is about 950°C. This sets a limit to the efficiency. Because the steam cycle provides approximately 80% of the power output, the PFBC efficiency depends mainly on the steam conditions used. Plants currently in service achieve efficiencies of up to 42% (Wu 2006). Other projects aim to achieve higher efficiencies by combusting the clean gaseous fuel in addition to the hot cleaned flue gas from the fluidised bed (supplementary combustion). The purpose of this is to raise the flue gas temperature to standard gas turbine inlet temperatures. For this purpose, either natural gas or a fuel gas produced by coal gasification can be used. The latter is shown in Fig. 7.10.

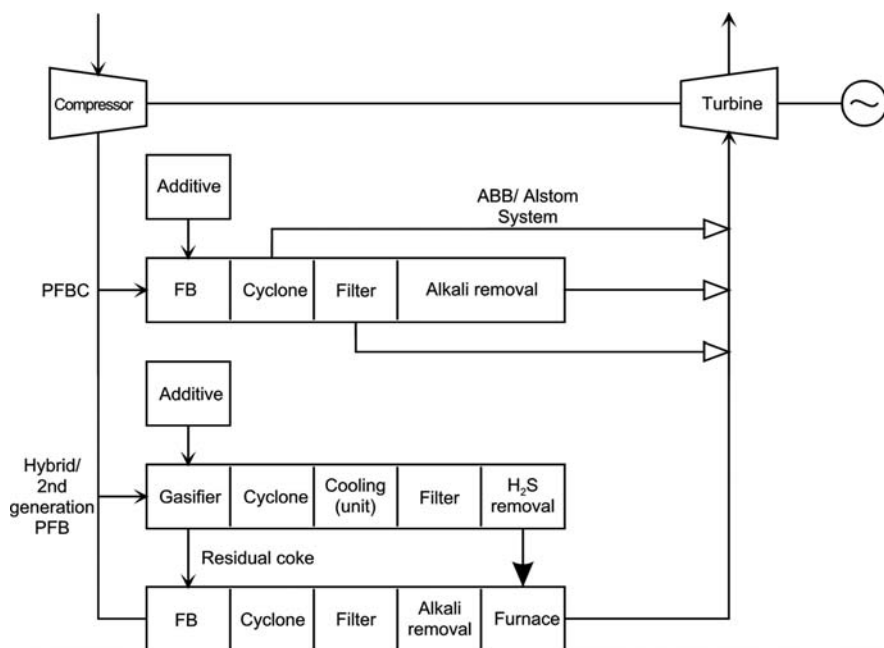


Fig. 7.10 Configurations of PFBC furnaces (Thambimuthu 1993)

For a hybrid or second-generation PFBC process of this nature, efficiencies of up to 52% are reported (when advanced gas turbine inlet temperatures and an advanced steam-production process are used). However, the higher gas turbine inlet temperatures generated by supplementary combustion place higher requirements on the gas cleaning stage after the PFB furnace. This in turn requires advanced filter systems and an alkali removal stage (Robertson et al. 2005).

A PFBC furnace is designed in such a way that the steam generator is installed inside a pressure vessel, because the compressive forces generated by the furnace pressure cannot be absorbed by the furnace's heat-absorbing enclosing walls. Because of the steam generator size and geometry, the vessels considered are cylindrical or spherical in shape.

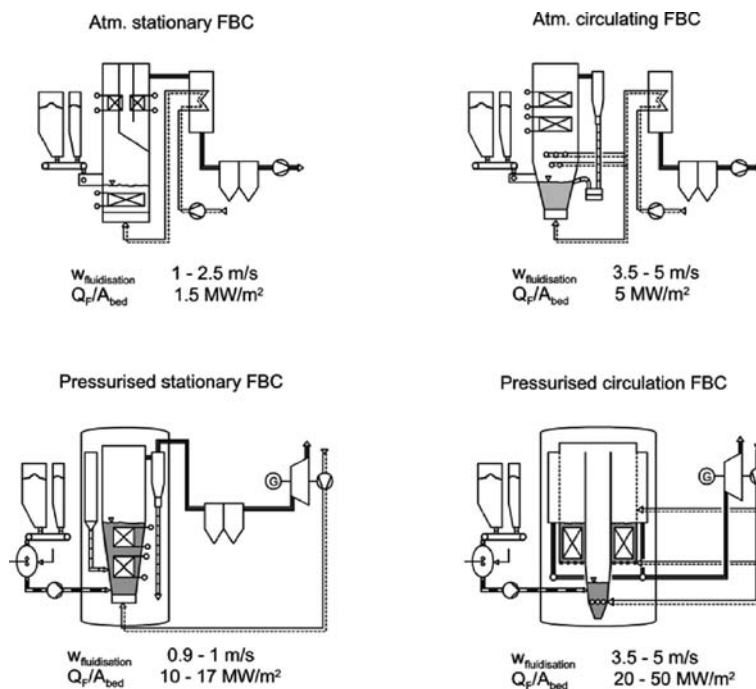
The pressurised design facilitates a much more compact furnace compared to atmospheric plants. The advantage of operating under pressure lies in the fact that the oxygen partial pressure of the combustion air increases as the operating pressure increases. As a result, the fuel throughput can be considerably higher for a given steam generator size.

The cross-sectional heat release rate  $\dot{q}_f/a_{\text{bed}}$  specifies the specific thermal output in relation to the bed surface. It is therefore a factor in determining the size of the fluidised bed combustor. Assuming equal fluidising velocities, the cross-sectional area heat release rate increases in proportion to the pressure (Bunthoff and Meier 1987).

In Fig. 7.11, the bubbling (stationary) and the circulating FBC types are compared with each other, with and without pressure. The parameters used for comparison are the superficial velocity and the cross-sectional heat release rate.

Atmospheric bubbling firing has fluidising velocities between 1 and 2.5 m/s. The speed cannot be further increased as it could cause erosion of the in-bed heat exchange surfaces. Due to this erosion hazard and the higher level of emissions of CO and NO<sub>x</sub> in bubbling FBC furnaces, circulating FBC has become the usual technology for plants operated at atmospheric pressure. Here, the fluidising speed can be raised to 8 m/s, because there are no heat transfer surfaces inside the fluidised bed. As a result, the cross-sectional heat release rate in atmospheric combustion plants rises from 1.5 MW/m<sup>2</sup> in stationary operation to 5–7 MW/m<sup>2</sup> in circulating operation.

Pressurisation allows the plant capacity to be increased considerably. In order to prevent erosion of the heating surfaces, the fluidising speed is limited to 1 m/s for bubbling pressurised fluidised beds. Despite this, the cross-sectional area heat release rate is between 10 and 17 MW/m<sup>2</sup> because of the increase in pressure. If this combustion technology is changed to a circulating type of pressurised fluidised bed, cross-sectional heat release rates of up to 50 MW/m<sup>2</sup> can be achieved. One outcome of this is a relatively slim pressure vessel design. The bubbling and the circulating FBC furnaces thus differ in geometry or, more precisely, in the proportion of the bed cross-section to the boiler height. Circulating FBC furnaces have a slimmer body and also promise lower emission levels. However, a circulating type features a more complicated plant design (JBDT 1992).



**Fig. 7.11** Comparison of bubbling (stationary) and circulating fluidised beds with and without pressure (JBTD 1992)

Although pressurised circulating FBC furnaces offer a range of advantages, only stationary combustion types are used commercially. Pressurised circulating FBC furnaces have been developed in Germany and Finland (Renz 1994). A pilot pressurised circulating fluidised bed furnace has been tested by Foster Wheeler as one component in a second-generation PFBC (Wheeldon et al. 2001). Figure 7.12 shows the commercial plants which have been built to date and their electrical capacities. Most of the pressurised FBC furnaces which have been planned and built are in Japan (McMullan 2004; Schemenau and van den Bergh 1993; Wu 2006).

In designing a fluidised bed, complete combustion must be ensured either through a sufficient residence time in the furnace or by ash recirculation. The particles emitted from the bed of a stationary FBC are completely combusted by means of higher combustion reaction velocities at a higher pressure. The size of the emitted particles depends on the fluidising speed and the density of the particles. Under the conditions offered by a bubbling fluidised bed with a fluidising speed of about 0.9 m/s, the emitted particles are smaller than 250  $\mu\text{m}$ . Since these particles are completely combusted in the freeboard volume of the furnace, they do not have to be recirculated under pressurised fluidised bed conditions. As a result of the higher fluidising velocities of circulating combustion, coarser particles are also emitted. This means

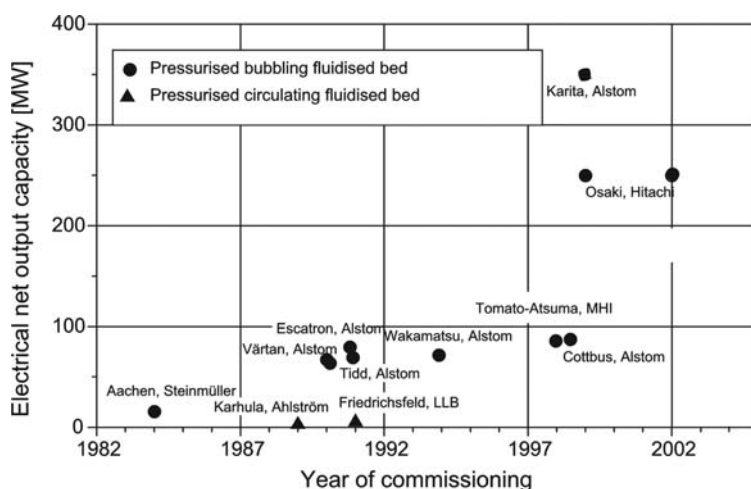


Fig. 7.12 Commercial pressurised FBC furnaces (data from Wu 2006; Schemenau 1993)

that the particle residence time in the freeboard stays below the necessary burnout time, and hence complete burnout is only achieved after several recirculations.

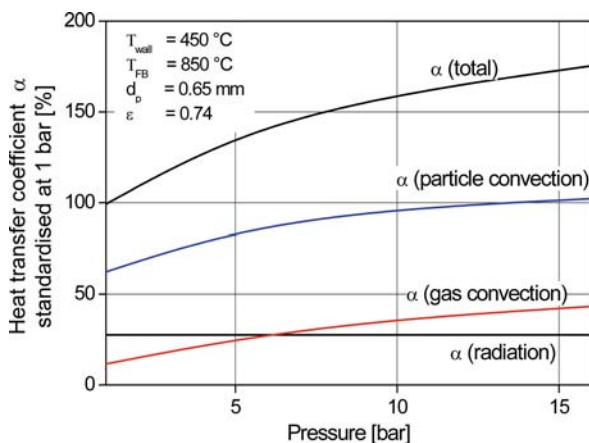
In order to reduce sulphur dioxide emissions, limestone is added to the combustion. It binds the developing sulphur dioxide and forms calcium sulphate. Atmospheric fluidised beds require ash recirculation in order to make better use of the limestone and to improve the sulphur capture. Recirculation is not needed in pressurised FBCs because of the better reaction kinetics of desulphurisation under pressure (Bunthoff et al. 1990).

In bubbling atmospheric FBCs, the bed height, generally around 1 m, is limited by the bed's pressure loss. An excessive loss should not be chosen for economic reasons. In comparison, the high thermal load and the large heat extraction surface of the bubbling PFBC furnace require higher beds of between 3 and 6 m. It is possible to use these high beds because the relative pressure loss, in relation to the process pressure, is low (Bunthoff and Meier 1987).

Pressure has a positive effect on the heat transfer. The heat transfer coefficients depend on the grains of the bed material, the fluidising velocity and the tube wall temperatures. The relationship between the pressure and the heat transfer coefficient is shown in Fig. 7.13. This figure shows that, compared to atmospheric operation, an increase in pressure to 16 bar (through the increase in gas density and the dynamic viscosity) improves the heat transfer of a bubbling fluidised bed by about 80%. Consequently, smaller heat transfer surfaces can be built for the same thermal output (Bunthoff and Meier 1987).

Oscillations of the grains of the fuel influence the heat absorbed by the in-bed heat transfer surfaces of the bubbling fluidised bed. An in-bed heat transfer surface which is large enough for one fuel type may be too large or too small for another fuel and may affect both the steam generation and the gas turbine inlet temperature. The

**Fig. 7.13** Effect of pressure on heat transfer in a pressurised fluidised bed (Bunthoff and Meier 1987)



division between coal combustion and heat dissipation in a circulating FBC process avoids these problems (JBDT 1992).

The design of a PFBC plant is determined by the gas turbine chosen. The compressor supplies the air mass flow required for fluidisation, combustion and cooling of the pressure vessel. It thus determines the boiler's capacity. The operating regime of the gas turbine at part load is determined by the need to set as large as possible a constant air excess level for the fluidised bed over the part-load range. This causes the pressure and the air mass flow to the fluidised bed to diminish in proportion to the thermal output (Stuhlmüller et al. 1995).

When used as a fuel, natural gas has a ratio of flue gas to air mass flow of about 1.02. In contrast, wet coal paste feeding gives between 1.07 and 1.11 and can rise to 1.3 with run-of-mine brown coal. Accordingly, the compressor of a gas turbine designed for natural gas would be too big or the turbine too small for wet coal paste FBC.

Large, stationary gas turbines are usually single shaft. The compressor, gas turbine and generator have a common shaft, and the revolutions per minute are determined by the power frequency. As a result, they cannot be used for load control (of the mass flow). For natural gas, it is possible to vary the mass flow by controlling the inlet vane for loads slightly above 50%; for smaller loads, the turbine inlet temperature is decreased. If inlet vane control is used in pressurised FBC to adjust the smaller compressor mass flow, the possible inlet vane control range is limited during part load. Below an inlet vane benchmark (which lies at about 70% of the rated useful heat output for hard coal and at about 80% for brown coal), single-shaft turbines make it necessary to bypass the fluidised bed in order to keep the excess air in the combustion at a constant level. However, the mixing of hot flue gas with the cold compressor air results in a decreased efficiency.

In comparison to single-shaft turbines, dual-shaft turbines allow better adaptation of the mass flow and the compressor pressure to the conditions of the pressurised fluidised bed, because the compressor's revolutions can be set independent

of the grid's frequency. With two exceptions, all existing plants with bubbling PFBC implement dual-shaft turbines of the same design. However, modern stationary gas turbines from all manufacturers, without exception, are currently single-shaft turbines (Stuhlmüller and Schauenburg 2001).

Combined cycle processes with pressurised fluidised bed combustion also differ from the natural gas fuelled combined cycle due to the much higher pressure losses in the fluidised bed boiler, filtering system and pipework – about 1 bar in total as opposed to 0.3 bar with natural gas. The pressure losses diminish the turbine's output. Since the impact on the efficiency is limited, the loss in efficiency is regarded as not too serious, and the fact that adapting the compressor blading is highly complicated further discourages addressing this drawback.

The gas turbine inlet has a low temperature of around 850°C. This is significantly lower than the temperatures of gas turbines in use today. With these lower temperatures, blade cooling can be simplified or dispensed with, meaning less cooling air is required.

### ***7.3.2 Hot Gas Cleaning After the Pressurised Fluidised Bed***

Hot gas cleaning after fluidised bed combustion concentrates on particulate matter removal (fly ash), although in some plants alkali removal may be taken into consideration as well (see Sect. 7.4.3). Sulphur is captured in the fluidised bed, while nitrogen oxide formation can be limited by the conditions of the combustion.

FB combustion has a temperature of up to 900°C, which is well below the ash deformation temperature. As a result, the separation process has to remove solid particulate matter from the hot gas flow. Various technologies for hot gas fly ash removal in PFBC furnaces are discussed below. They are

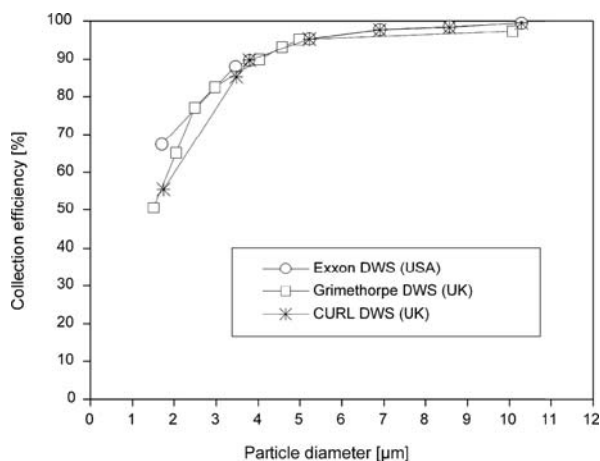
- cyclone separators,
- electrostatic precipitators (ESPs) and
- particulate collectors such as candles, tubulars, bags or cross-flow filters and packed-bed filters.

#### **7.3.2.1 Cyclone Separators**

Cyclone separators use the principle of separation by centrifugation. The flue gas, laden with fly ash, enters the cyclone tangentially, creating a downward rotating flow, changing direction as it reaches the bottom of the cyclone. It then leaves the cyclone at the top, free from fly ash (see also Fig. 5.76). The ash particles are pushed outwards by centrifugal force, hit the walls and fall by gravity to the bottom of the cyclone, where they are removed.

Cyclone separators are simple in design and feature high throughputs and removal rates. The removal rate depends on several factors. The higher the inlet speed, particle size and density and the smaller the cyclone diameters, the greater the centrifugal

**Fig. 7.14** Cyclone collection efficiency as a function of particle diameter (Thambimuthu 1993)



force and hence the removal rate of a cyclone. The removal rate diminishes at higher temperatures due to the increasing gas viscosity (Thambimuthu 1993). Reported total removal rates under pressurised fluidised bed conditions range from 83 to 98% with one-stage and from 98 to 99.6% with multistage cyclone separators (Schiffer 1989).

A cyclone's efficiency also depends on the particle size. It decreases considerably for particles between 5 and 10  $\mu\text{m}$ . Figure 7.14 shows the relationship between the removal rate and particle size as determined by measurement after FBC furnaces in experimental facilities (Thambimuthu 1993). Increasing the removal rate for small particles by means of changing design parameters and operating conditions has only limited effectiveness, because acceleration of the flue gas speed results in increased pressure losses and erosion. A pressure loss of 0.8 bar is assumed for a two-stage cyclone separator after an industrial-scale pressurised FBC furnace (Mustonen et al. 1991).

Single-stage cyclones are used for pre-separation; two or three series-connected cyclones are used for final fly ash separation. State-of-the-art ash separation in the pressurised FBC furnaces currently in use is performed by two-stage cyclone separators. Given a removal rate of 99%, the flue gas shows a cleaned gas particulate concentration of 200–500 mg/kg before entering the turbine, with a maximum particle size of 10  $\mu\text{m}$  (Jansson 1995b). In this case, the gas turbine features a modified design in which the turbine parts subject to erosion have a high wear resistance to the remaining fly ash in the gas.

Cyclone separators are always designed for a defined volumetric flow. If the flow, and hence the inlet speed, decreases, the removal rate also falls. The volumetric flue gas flow should therefore be kept constant in order to ensure a sufficient removal rate, even under partial-load conditions.

### 7.3.2.2 Electrostatic Precipitators

An electrostatic precipitator (ESP), used to remove particulate matter, is a well-established piece of technology in coal-fuelled power plants. The principles of ESPs have been discussed in Sect. 5.9.2 in the context of pulverised coal combustion. For pulverised coal combustion, the ESP is located at the cold end of the flue gas path in the temperature range of about 150°C. In the temperature and pressure range of pressurised fluidised bed combustion, the removal behaviour of ESPs has only been investigated in laboratories. These investigations revealed a remarkably high ESP energy demand, which has to be attributed to the lower electrical resistance of fly ash at higher gas temperatures.

Although negative results, which would exclude the principle of the use of an ESP under pressurised FBC conditions, have not been reported, there is no information available about R&D projects which continue the work in this field (Weber et al. 1993; Takahashi et al. 1995; Renz 1993).

### 7.3.2.3 Filtration Separators

Filtering separators use filter media such as granular bulk materials, sintered ceramic material, tissue, felt, non-woven material or shaped pieces of fibre. Particulates deposit on the surface of these and are cleaned off periodically. For fly ash collection in pressurised fluidised beds, packed filter beds made from temperature-resistant granular material, ceramic filter media in the form of bags, and rigid elements in the form of plates or cylinders are the types of equipment investigated today (Wu 2006; Jansson et al. 1996; Sasatu et al. 2001; Toriyama et al. 1999; Newby et al. 2001; Newby et al. 1999; Weitzel and McDonald 1999; Wheeldon et al. 2001; Wright et al. 2003).

#### – *Packed-bed or granular bed filters*

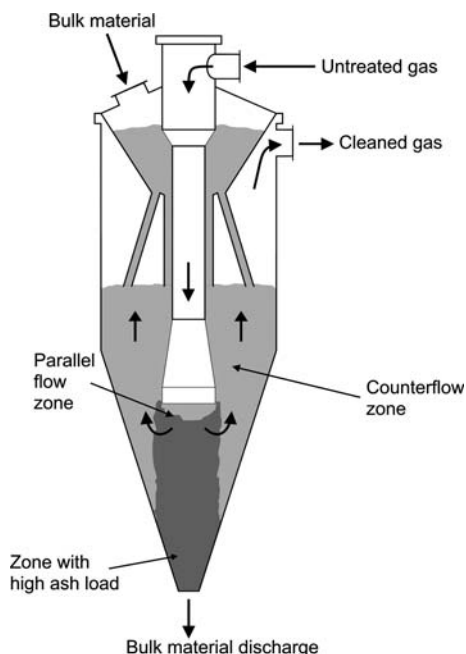
In a packed-bed filter, the gas containing the particulates flows through a bulk bed made of granular material, where it deposits the particles. Figure 7.15 shows such a packed-bed filter. The untreated gas containing the particulates from combustion flows into the bed through a concentric duct from above. It is deflected inside the bulk bed and extracted in the form of cleaned gas at the top. As the particulate deposits accumulate in the bulk bed, the bed's flow resistance increases. The polluted bulk material is continuously discharged at the bottom while fresh bulk material is supplied from above.

Today, packed-bed filters are used to collect particulate matter contained in hot waste gases from industrial furnaces containing abrasive, chemically aggressive and/or sticky dusts. Investigations into the sorption of gaseous pollutants show that packed-bed filters can also be used to remove pollutant gases such as HCl, SO<sub>2</sub> and alkalis (Schiffer 1989).

Packed-bed filters are efficient at removing particulates from hot gases and satisfy the purity requirements of gas turbines. During investigations under FBC conditions, removal rates of 97–99% and a particulate concentration of



**Fig. 7.15** Schematic drawing of a packed-bed filter (Thambimuthu 1993)



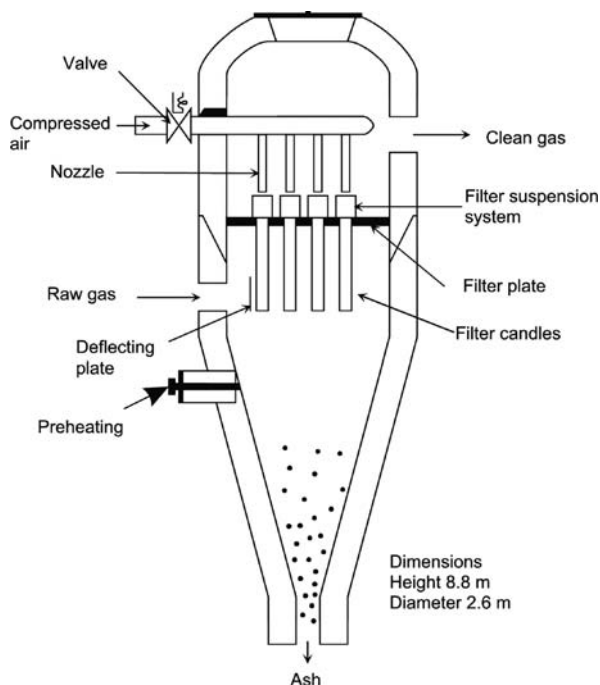
4–6 mg/m<sup>3</sup> in the cleaned gas were achieved, with pressure losses of 50–80 mbar. The flow velocities range from 0.1 to 0.3 m/s (Renz 1993). Compared to cyclone separators, it is clear that less pressure is lost when packed-bed filters are used. Even if a cyclone is installed upstream, the total pressure loss reported is about 200 mbar. This is also lower than the rate of the two-stage cyclone (Mustonen et al. 1991).

A packed-bed filter installation needs considerably more equipment, however. This includes vessels for cleaning and for the polluted bulk material, the appropriate lock-hopper system for pressurised operation and a regeneration system for the polluted bulk material (Mustonen et al. 1991).

#### – *Candle filters*

Candle filters are named after their shape, which resembles a hose with a closed end. Figure 7.16 shows the installation of a candle filter in a pressure vessel, while Fig. 7.18 shows the design for a 150 MW<sub>el</sub> PFBC plant. The untreated flue gas containing the particulates enters the lower part of the vessel, flows through the filter candles and is extracted in the upper part of the vessel. The filter candles are held in place both vertically and horizontally by a perforated plate. The untreated gas and cleaned gas zones are separated from each other by the candles and plate. The filter medium is porous and only allows very small fly ash particles to pass through. The maximum size depends on the pore diameter of the filter material. A filter cake forms on the outside of the filter candle and plays a major part in fly ash collection and also increases the pressure loss. The filter cake can be removed by

**Fig. 7.16** Schematic drawing of a candle filter (Thambimuthu 1993)



means of compressed air pulses from the side of the cleaned gas. The particulates are extracted from the bottom of the vessel.

The candle tubes, closed at their bottom end, are made of a coarsely porous sintered ceramic material. Typical filter elements have an outer diameter of 60 mm, a wall thickness of 10–20 mm and a length of 1–1.5 m. If the material is silicon carbide, the pore diameter ranges between 30 and 100  $\mu\text{m}$ . The filter efficiency can be improved by applying a finely porous surface layer of ceramic fibres to the side of the ceramic material facing the untreated flue gas. These fibres are firmly joined by sintering with the main body. When a two-component system such as this is used, the pore sizes can be as small as 3  $\mu\text{m}$  (Thambimuthu 1993).

The filter shown in Fig. 7.16 was installed after a cyclone separator for long-term tests at an experimental plant. Given an initial particle concentration of 1,000–4,000 mg/kg in the untreated gas, with an average particle diameter of 3–7  $\mu\text{m}$ , removal rates of 99% were reached. This meant that particulate concentrations below 10 mg/kg were achieved in the cleaned gas. The approach speed at the filter candles ranged between 0.03 and 0.07 m/s (Thambimuthu 1993). At an approach speed of 0.07 m/s, the pressure loss was 230 mbar. The basic disadvantage of the configuration shown in Fig. 7.16 is that filter suspension and the cleaning system cannot be scaled up for larger filter units (Renz 1993).

The only practical method for cleaning the filter candles while ensuring a constant flue gas mass flow to the gas turbine is pulse jet cleaning. A method

involving constant switching of single filter modules would not be compatible with the operating regime of the gas turbine. What is more, there are no sufficiently reliable hot gas valves available for this purpose (Stuhlmüller et al. 1995).

Even though ceramic candle filters are highly developed, their suitability for use in industry has not yet been proven. At temperatures above  $775^{\circ}\text{C}$ , it gradually becomes more difficult to remove the filter cake as ash properties begin to change and as the operating pressure loss increases (Jansson and Svensson 1997). In different PFBC demonstration projects, ceramic filters have been tested using a partial flow of hot flue gas. At the Tidd plant, ceramic candle filters were installed after one of the primary cyclones. The main problems encountered in the test included ash bridging between candles, difficulties in cleaning the candles and difficulties in draining the filter vessel. The bridging reduced the effective filtration area and, more seriously, led to mechanical failure of the elements under tensile stress. At the Escatrón plant, ceramic candle filters were tested at  $750\text{--}820^{\circ}\text{C}$ . The filters were also subjected to ash bridging and mechanical failure (Wu 2006; Wright et al. 2003).

– *Bag filters*

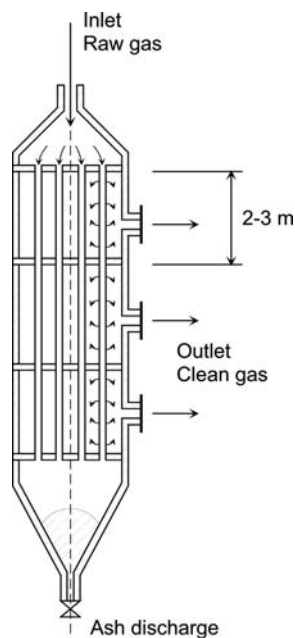
In contrast to the self-supporting candle filters, bag filters need a support cage to maintain the form of a hose because of their fabric filter. Ceramic or metallic materials are the fabrics considered for the application of bag filters in the temperature range of FBC furnaces. A survey of filter materials which have already been investigated can be found in Thambimuthu (1993).

– *Tube filters*

Tube filters work according to the same principle as candle filters. The untreated gas, however, approaches from inside the tubes. The design of a tube filter is shown in Fig. 7.17. The gas flow containing the particulates enters the vessel and ceramic tubes from above and exits as clean gas. The particulates are removed as the flue gas radially passes through filter tube walls. The particulate matter collects inside the tube, is removed by compressed air, pushed downwards, collected at the vessel bottom and discharged. The tubes have large internal diameters of 140 mm in order to prevent clogging of the filter media. The tube is up to 6 m in length, which gives the advantage of a compact design (Thambimuthu 1993).

This configuration was used in a circulating PFBC test furnace of  $10\text{ MW}_{\text{th}}$  and in a stationary PFBC test furnace of  $15\text{ MW}_{\text{th}}$ . An outlet particulate load of around  $3\text{ mg/Nm}^3$  was achieved in the cleaned gas. At the  $71\text{ MW}_{\text{el}}$  stationary PFBC furnace in Wakamatsu, Japan, the tube filter technique was tested during demonstrative operation (Sasatu et al. 2001). Following a coarse separation stage in cyclones, the whole flue gas flow was cleaned in two tube filters. Each tube filter was 3.2 m in diameter and 16 m in height. Under trouble-free operation conditions, the design parameters of about  $2\text{ mg/Nm}^3$  for the particulate matter content and 100 mbar for the pressure loss were achieved. Tests were carried out in two phases, with a total of 11,500 h of operation. The longest surviving filter had a lifetime of approximately 8,000 h (Wu 2006). On the whole, however, a series of problems such as filter failure and ash clogging occurred.

**Fig. 7.17** Schematic drawing of a tube filter by Asahi Glass, Japan (Thambimuthu 1993)

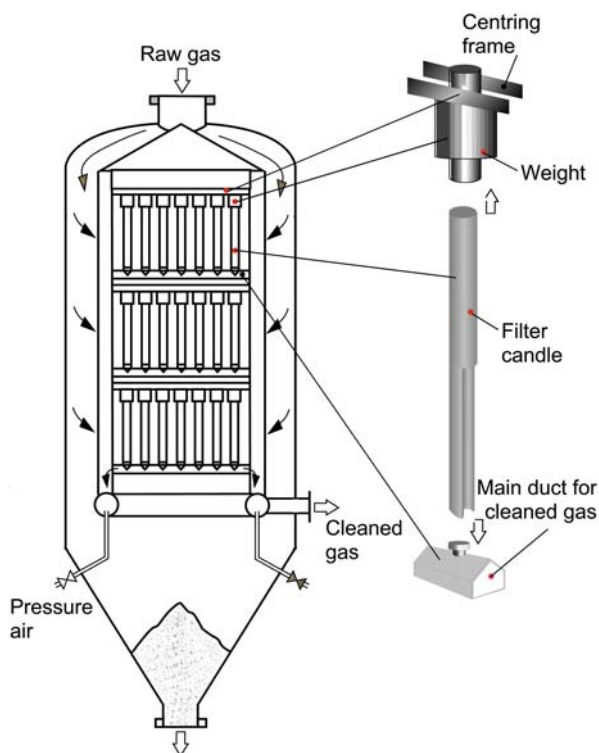


#### 7.3.2.4 Comparison of Methods and Techniques

In combined processes with pressurised FBC, the removal of particulate matter such as fly ash from the hot gas is an essential component in the production of hot gas suitable for a gas turbine. In addition to cyclone separators currently in use, other methods are being developed and demonstrated.

Cyclones for fly ash removal are characterised by both high velocities and high pressure losses (Thambimuthu 1993). It is possible to achieve removal rates of 99% using two cyclones, though the requirements of modern gas turbines are not met by doing so. However, cyclone separators are nevertheless used in PFBC furnaces because proof of the operating reliability of the other techniques has not yet been established. Ceramic candle filters work with low approach velocities of less than 0.04 m/s, involving low pressure losses of about 200 mbar. These filters are the most advanced of the various filtration separation methods described above.

Based on several years of investigations, the candle filter technique has been demonstrated at various industrial-scale plants. Tube filters are almost as highly developed as ceramic candle filters. Although they exhibit an acceptable filtration performance in demonstration plants, ceramic candle and tube filters are not yet sufficiently reliable for the advanced power generation demonstration plants. Their unreliability makes operation uneconomic. Thus the continuing development of hot gas filter technology is focussed on improving reliability (Newby et al. 2001). Given the high mechanical and thermal stresses, to attain greater reliability, the filter elements and the material and better constructions and configurations have been highlighted as potential areas for development. Ceramic-based bag filters are clearly not as highly developed as candle filters.



**Fig. 7.18** Candle filter of a 150 MW<sub>el</sub> power plant with circulating PFBC furnace (Bauer et al. 1994; Rehwinkel et al. 1992)

Figure 7.18 shows the design of a ceramic candle filter for a planned 150 MW<sub>el</sub> power plant with a circulating PFBC furnace (Bauer et al. 1994; Rehwinkel et al. 1992). The flue gas containing the particulates is fed into the filter vessel and candles from above. An impact plate acts as a flow deflector, ensuring a steady flow of solid flue gas across the entire cross-section of the filter.

In the filter unit shown, 1,800 ceramic filter candles are arranged upright on supporting headers on three levels. This vertical mounting has several advantages compared to the suspended mounting. First, the weight load on the ceramic material takes the form of compressive stress and not tensile strain, which is the preferable design for ceramics. Furthermore, the filter candles, with an increasing degree of pollution, are pressed more firmly into their sealed fit. The concentration of outlet particulates in the cleaned gas is less than 5 mg/Nm<sup>3</sup> (Bauer et al. 1994).

Packed-bed filters work at higher velocities (0.2 m/s) and have lower pressure losses of less than 200 mbar. In addition, they allow the removal of other substances which are likely to damage the gas turbine. Although packed-bed filters are not yet as highly developed as candle filters, they are expected to be just as suitable as them in general.

### ***7.3.3 Pressurised Bubbling Fluidised Bed Combustion (PBFBC)***

#### **7.3.3.1 State of Development**

The technology used in bubbling pressurised fluidised bed combustion is highly advanced. A total of eight PBFBC plants have been built around the world, giving a cumulative installed capacity of 1,125 MW<sub>el</sub> to date (McMullan 2004; Wright et al. 2003; Wu 2006). Some of the plants initially functioned as demonstration units; today, however, the majority are operated on a commercial basis. The company known as ABB Carbon, now part of Alstom Power, has supplied most of the installations; two units have been built in Japan with Japanese PFBC technology. Overall, the uptake of PBFBC technology has been progressing slowly. Table 7.6 summarises the details of existing PBFBC plants.

Four demonstration plants were built around 1990. They were based on ABB Carbon's P200 module and had a thermal output of about 200 MW<sub>th</sub>. Two of them are operated by Stockholm Energi in Värtan, Stockholm, Sweden, and one by ENDESA in Escatrón, Zaragoza, Spain. The Tidd plant near Brilliant, Ohio, operated by American Electric Power, was taken out of service in 1995 after operating as a demonstration plant for several years. The Tidd and Escatrón plants were designed for an electrical output of 70 and 76 MW<sub>el</sub>, respectively, whereas the Värtan plant (two P200 modules) is operated as a CHP power station with an electrical output of 135 MW<sub>el</sub> and a thermal heat extraction rate of 225 MW. In Escatrón, hot gas filtration using a partial flue gas flow was investigated (Jansson 1995a; Jansson et al. 1996; Jansson 1995b; Martinez Crespo 1995).

In 1993, another plant entered service in Kyushu, Japan. This plant is also equipped with a filtration separator for the whole flue gas flow (Goto 1995; Sasatu et al. 2001). In 1999, a pressurised fluidised bed based on the P200 module and fuelled by brown coal entered service in Cottbus, Brandenburg, Germany (Walter et al. 1997). Based on the experience with the P200 module, an FBC furnace was designed and manufactured for a plant in Karita, Japan. This furnace featured a thermal capacity of 800 MW<sub>th</sub>. The plant began demonstration operation in 1999 and entered commercial operation in 2001. It features advanced supercritical steam conditions (24.1 MPa/566°C/593°C) and a net thermal efficiency rate of nearly 42% (HHV), corresponding to about 44% (LHV) (Koike et al. 2003; Asai et al. 2004). This represents state-of-the-art PFBC technology (Jansson and Anderson 1999).

Other PBFBC plants have been built by Japanese companies in Japan: the 85 MW<sub>el</sub> Tomatouatsuma Unit No. 3 built by Mitsubishi Heavy Industries (MHI), which uses a ceramic filter, and the 250 MW<sub>el</sub> Osaki Plant by Hitachi (Shimuzu and Itoh 2001; Hokari et al. 2001).

The apparent lack of market penetration in Europe and North America is believed to be a result of PBFBC's perceived higher costs and complexity compared to competing systems, coupled with an increasing focus on natural gas fired combined cycle gas turbine plants. As Japan does not have access to substantial gas reserves and is a net importer of LNG, the future market potential is seen as being mainly in Japan. Due to rationalisation of Alstom resources, PBFBC is no longer actively

**Table 7.6** Summary data for PBFBC plants currently in service (data from Wu 2006 and additions)

Project/typical feature	Värtan	Escatrón	Tidd	Wakamatsu	Cottbus	Tomatouatsuma	Osaki	Karita
Start-up date	1989	1991	1991	1994	1998	1999	1999	1999
Type	ABB 2xP200 CHP	ABB P200 Cond.	ABB P200 Cond.	ABB P200 Cond.	ABB P200 CHP	MHI	Hitiachi	ABB P800 Cond.
$P_{el}$ , net	135.0 MW	79.5 MW	70.5 MW	71.0 MW	74 MW	85 MW	250 MW	360 MW
$P_{th}$	224.0 MW	—	—	—	120 MW	—	—	—
Gas turbine	2xGT35P	GT35P	GT35P	GT35P	GT35P	MW-15 IP	GE F7EA	GT140P
Boiler pressure	12 bar	12 bar	12 bar	12 bar	12 bar	12 bar	10 bar	12 bar
Bed temp.	860°C	860°C	860°C	860°C	840°C	870°C	865°C	870°C
Gas clean-up	Cyclones	Cyclones	Cyclones	Cyclones + ceramic filter	Cyclones	Cyclones + ceramic filter	Cyclones	Cyclones
Coal	Hard coal, Poland	Brown coal	Hard coal, Ohio	Hard coal	Brown coal, dried	Brown coal, dried	Hard coal	Hard coal
Sulphur <sup>a</sup>	0.1–1.5%	2.9–9.0%	3.4–4%	0.3–1.2%	< 0.8%	< 0.8%	—	0.3–1.2%
Ash <sup>a</sup>	8–21%	23–47%	12–20%	2–18%	5–6%	5–6%	—	2–18%
Moisture <sup>a</sup>	6–15%	14–20%	5–15%	8–26%	18–20%	—	—	8–30%
Coal feeding	Paste	Dry	Paste	Paste	Dry	—	Paste	Paste
Sorbent feeding	Paste with coal	Dry	Dry	Paste with coal	Dry	Dry	Paste with coal	Paste with coal
Sorbent	Dolomite	Limestone	Dolomite	Limestone	Limestone	—	Limestone	coal
Live steam pressure	137 bar	94 bar	90 bar	103 bar	142 bar	166 bar	167 bar	Limestone
Live steam temp.	530°C	513°C	496°C	593°C/593°C	537°C/537°C	566°C/538°C	571°C/596°C	570°C/595°C
Bed height	3.5 m	3.5 m	3.5 m	3.5 m	—	—	4 m	3.5 m
Net efficiency, %	—	36 (HHV)	35 (HHV)	38 (HHV)	—	41 (HHV)	42 (HHV)	44 (LHV)

<sup>a</sup> As-mined coal

marketed, although support is still provided for existing installations. It seems that the PBFBC technology will only flourish if its use is promoted by the Japanese licensees and their local competitors or if it is utilised in some form of a hybrid cycle (McMullan 2004).

### 7.3.3.2 Industrial-Scale Configurations

The design of ABB Carbon's P200 module will be described in more detail below, as it is the most widely used PFBC technology and information about its design and operation has been published. Figure 7.19 shows the plant in Cottbus, which is based on pre-dried brown coal (Walter et al. 1997; Jansson and Anderson 1999). It can be divided into three sections – a gas turbine cycle, a steam – water cycle and a fluidised bed.

The combustion air is taken in from the ambient air, pre-compressed in a low-pressure compressor (LP compressor), intercooled and then further compressed to reach the required working pressure for combustion. Intercooling is chosen so that low inlet temperatures of around 300°C can be set in order to cool the pressure vessel. Afterwards, the air is injected into the furnace via the distributor plate.

The flue gas produced by combustion is cleaned in a two-stage cyclone separator. It is then directed, in the form of hot gas, to the high-pressure gas turbine via a coaxial duct, inside which the hot gas flows, while the compressed air flows on the outside. In the high-pressure gas turbine, the hot flue gas is pre-expanded, then further expanded in the low-pressure gas turbine, in order to drive the low-pressure compressor.

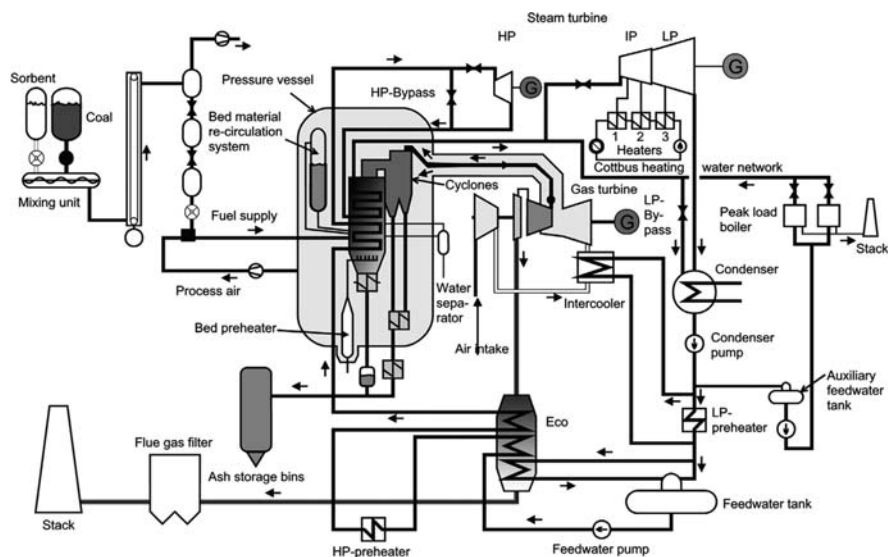


Fig. 7.19 Diagram of the PBFBC power plant in Cottbus (Walter et al. 1997)



The low-pressure cycle, featuring an LP gas turbine and a pre-compressor, is driven at variable speed; the high-pressure cycle, featuring a HP turbine, a HP compressor and a generator, is driven at constant speed. Power output is generated exclusively by the HP gas turbine, which drives the generator via gears. After passing through the two cyclone separators, the particulate matter typically comes to between 200 and 500 mg/kg (Jansson 1995b), with a maximum particle size of about 10  $\mu\text{m}$ . The gas turbine used has been modified and adapted in order to suit the cleaned hot gas from the PFB – to limit the impact of erosion, corrosion and fouling and to achieve a longer lifetime.

A standard gas turbine was modified for low flow velocities by increasing the number of stages (in order to reduce the deflections) and by reinforcing the blade profiles of the parts at risk. This made it more suitable for operation after the fluidised bed combustion furnace. With its pressure conditions and mass flow, the gas turbine chosen determines the design of the pressurised FBC furnace. Since it is too expensive to develop a gas turbine especially for pressurised FBC furnaces, recourse is made to suitably modified existing gas turbines. The compressor mass flow then sets the key values for the design of such a furnace.

The waste heat from the gas turbine can only be used for feed water heating at relatively low gas turbine outlet temperatures in the steam – water cycle. This heat accounts for 10% of the steam produced. The far greater portion of heat in the steam produced in fluidised bed combustion – about 90% – is transferred directly to in-bed heating surfaces and furnace walls and used for vaporisation and superheating. The gas turbine accounts for only a small share of the total power output. This is because the steam – water cooling of the fluidised bed (by means of in-bed heating surfaces, in order to maintain the FBC temperatures) has to stay below 850°C and because this heat is not used in the gas turbine. The output ratio of the gas turbine to the steam turbine is about 1–5.

The pressure vessel in the P200 module in the installed plants is 20 m high and has a diameter of 13 m. In these plants, the cyclones are located to the side of the fluidised bed component. In the Wakamatsu and Cottbus plants, the cyclones are located above the fluidised bed which, with a diameter of 11 m and a height of 32 m, makes the body of the pressure vessel higher and more slender.

The vessel interior is cooled by the combustion air coming from the compressor before it is used for fluidisation and combustion in the fluidised bed. Because of the low temperature, low-alloy steel types can be used for the pressure vessel.

In order to be used in FBC, the coal has to be ground to a grain size of less than 5 mm. Any further preparation depends on the calorific value and the sulphur content of the coal. In the case of the Värtan power station, coal and limestone are mixed with water and pumped into the firing facility by means of a slurry pump. The water content in this coal – water suspension amounts to about 25%. From a process-engineering perspective, wet feeding is less complicated than dry feeding using pneumatic conveying and a lock-hopper system. Coals with higher calorific values, ash contents below 25% and lower sulphur contents require less additives for desulphurisation. If such coals are used, losses in efficiency due to the necessary evaporation heat and the increased volumetric flue gas flow are compensated for by

the lower auxiliary power requirement of wet feeding. Wet feeding works without coal drying and pressurisation of the lock-hopper system.

Dry feeding is used for coals with a low calorific value and a high sulphur content, as in the Escatrón plant, because if wet feeding were used the high fuel and additive mass flows in this plant would lead to a reduced efficiency. In the Tidd plant, the fuel is fed in wet, while the additive for desulphurisation is dry.

A high fluidised bed of 4 m and a relatively low flow speed result in a sufficient residence time of about 4 s for the gas in the fluidised bed; the residence time in the freeboard above the bed is also around 4 s. The relatively long residence times, as well as the pressure conditions, favour burnout, resulting in a low carbon level of about 1% in the fluidised bed. This is the same as the level of unburned matter in the extracted ash. It is not necessary to recirculate the ash in order to raise the burnout in PFBC (Schemenau 1993).

### 7.3.3.3 Control

At partial load, the operating pressure is reduced to the level of the fluidisation speed and the inlet velocities to that of the cyclones, and the excess air has to be kept constant. The operating pressure and volumetric air flow are set by the variable-speed LP compressor in the cross-compound gas turbine, and the HP compressor unit is driven at a constant speed. The speed of the LP compressor is controlled by adjustable inlet guide vanes in the LP turbine (Keppel 1995). The power output of the gas turbine diminishes with a diminishing pressure.

Partial loads in the steam generator and steam turbine are set by means of the fluidised bed height. For this, the heating surfaces are only partly submerged into the fluidised bed, because heat generated by the gas is not transferred as effectively as that generated by the fluidised solid matter in the bed. As a result, heat absorption by the in-bed heating surfaces diminishes and steam production decreases. The flue gas cooling by the free-lying, in-bed heating surfaces (i.e. those portions of the heating surfaces that are not emerged in the bed) causes the gas turbine inlet temperatures to drop, resulting in a decreased efficiency.

So that the ash balance within the system is maintained during partial-load operation, the pressure vessel contains a buffer bed ash storage unit. As the load decreases, this unit removes bed ash from the fluidised bed and, in return, provides stored ash to refill the fluidised bed according to the (pre-determined) rated loads of ash. A pneumatic transport system stores the ash and discharges it. In the Cottbus plant, fuel oil is supplied to the firing unit at partial load in order to keep the freeboard temperature or the gas turbine inlet temperature constant across the entire load range. This also helps to reduce nitrogen oxide loads by means of ammonia injection before the cyclones (SNCR) (Jansson and Anderson 1999).

Experience at the Karita plant has shown that the minimum stable capacity is 40% of the nominal power output, and the load change rates are about 3%/min between a 40 and 90% load and 2%/min between a 90 and 100% load. A cold start from ignition to full load takes 11 h, a warm start takes 4 h and a hot start takes 3 h. When the plant is starting up, the compressor is first of all run by the generator,

which is operated as a motor, whereupon the fluidised bed is heated up by natural gas or fuel oil (Asai et al. 2004).

### 7.3.3.4 Emissions

Table 7.7 shows the emissions from selected PBFBC demonstration plants. Thanks to the low combustion temperature of the fluidised bed,  $\text{NO}_x$  emissions are generally lower than 200 ppm, but vary widely from plant to plant. As these plants use similar bed temperatures of around 860–870°C, this variation is probably due to the differences in the fuel-N content, fuel-volatile content and other parameters. The Värtan plant burns a bituminous coal with 1.3 wt% nitrogen and 27% volatile matter and has relatively high emissions of 165–191 ppm. At the Osaki plant, however, the uncontrolled  $\text{NO}_x$  emissions are as low as 14.4 ppm (Wu 2006). As the power output decreases, the  $\text{NO}_x$  emissions increase, because the heterogeneous reduction of the nitrogen oxides by the fluidised bed ash diminishes due to the lower bed height.

Compliance with emission limits requires further measures. In addition to injecting ammonia in order to control nitrogen oxide (selective non-catalytic reduction), manufacturers suggest a “freeboard” firing stage in order to increase the temperature and stage the fuel. Both methods have been applied in the Cottbus plant (Walter et al. 1997; Almhem 1996). In the case of high uncontrolled emissions, selective catalytic reduction (SCR) is the most effective, but also the most expensive,  $\text{NO}_x$  reduction technology. For example, the Karita plant has achieved  $\text{NO}_x$  emissions of 35–42 ppm by using SCR.

The plants can achieve desulphurisation of more than 90% by adding limestone or dolomite, depending on the Ca/S molar ratio. A sulphur retention of 90% is typically achieved at Ca/S molar ratios of 1.8–2.0. As shown in Table 7.7, the  $\text{SO}_2$  emissions for these plants vary widely from 5 to 350 ppm. The Värtan plant has particularly low  $\text{SO}_2$  emissions due to the low-sulphur (0.1–0.6%) coal burned and a high sulphur retention (96–98%). The Escatrón plant burns a high-sulphur (3–9%) coal but with a moderate sulphur retention (90–95%). This results in relatively high

**Table 7.7** Emissions from PBFBC plants in operation (Wu 2006)

Plant	Värtan	Tidd	Escatrón	Osaki	Karita
$\text{NO}_x$ without control, ppm	165–191	86–102	120–170	14.4	
$\text{NO}_x$ with control, ppm	20–33 (SNCR)				35–42 (SCR)
Sulphur retention, %	96–98	93	90–95	97.7	
Ca/S molar ratio	3.3	2.0–2.2	1.7–2.0		
Ca/S ratio at 90% sulphur retention	2.0	1.8–1.9	1.8		
$\text{SO}_2$ emissions, ppm	5–9		350	7.1	7–36
Particulates, mg/m <sup>3</sup>	< 30	18	76	≤ 3.5 (2-stage cyclones + bag filters)	5–15 (2-stage cyclones + ESP)

SO<sub>2</sub> emissions. The Värtan plant meets the German standard emission limits of 400 mg SO<sub>2</sub>/Nm<sup>3</sup> at 6% O<sub>2</sub>, whereas both Tidd and Escatrón exceed these standard limits by far. This is due to the high sulphur content of the coal used in these plants (Jansson et al. 1996).

Particulate emissions for the PBFBC plants in operation are also relatively low, between 3.5 and 76 mg/m<sup>3</sup>. The levels depend on the initial gas cleaning before the gas turbine (cyclones or ceramic filters) as well as final cleaning (ESP or bag filters) prior to discharge from the stack. For example, the Osaki plant employs two-stage cyclones for initial cleaning and bag filters for final cleaning. This plant has achieved particulate emissions as low as 3.5 mg/m<sup>3</sup> (Wu 2006).

### 7.3.3.5 Residual Material

In order to operate a power plant, it is essential to use, rather than dispose of, any residual material produced. In a PFBC furnace, residual material is produced in the cyclone filters (20–50%), the bag filter after the turbine (2%) and as a result of ash removal from the bed material (45–75%). According to manufacturers' data, the residual material mainly contains calcium sulphate (Ca<sub>2</sub>SO<sub>4</sub>), additive (CaCO<sub>3</sub>) and small quantities of quicklime (CaO), sulphites (CaSO<sub>3</sub>) and sulphides (CaS), in addition to coal ash. After the addition of water, the residual material is self-capturing, and eluted trace elements are scarce. Using the ash presents the same opportunities and problems as ash from atmospheric FBC furnaces (see Sect. 5.11). Examples of options for PFBC ash include using it as filling material in underground constructions or for road substructures (Rogbeck 1996).

### 7.3.3.6 Operating Expertise

The operational expertise gained at the different experimental plants is compiled and summarised in Stringer (1989). The major problems are erosion and corrosion of in-bed heating surfaces and gas turbines. Extensive investigations at the test plants revealed the need to improve the design of the in-bed heating surfaces, to choose lower flow velocities and to limit the grain size in order to minimise erosion of the surfaces. The corrosion phenomena of the in-bed heating surfaces can be compared to those in atmospheric FBC furnaces. Corrosion can be limited by the choice of material and by preventing air-deficient zones. An efficient particle removal technique is essential in preventing erosion of the turbine. The use of two-stage cyclone separators should help to meet these requirements, even though there is little leeway when there are disturbances. At FB temperatures below 850°C, problems are not expected to arise from corrosion of the gas turbine, though they are likely to occur at higher temperatures.

An overview of the operational expertise gained through use of the demonstration plants is summarised in Wright et al. (2003) and Wu (2006). A series of operational problems arose in all plants, as is quite common when new technologies are introduced for the first time. However, most of the problems were not associated with

pressurised fluidised bed combustion. The specific problems of PBFBC furnaces reported are discussed below.

### Erosion of In-Bed and Membrane Wall Heating Surfaces

The furnace walls feature a water-cooled membrane design and are lined with various refractories in order to prevent the bed materials from causing erosion. Heat transfer surfaces may be located within the furnace in the form of evaporator tubes, superheaters and reheaters. The evaporator tubes are typically made of low-grade steel with a 1 mm thick flame-sprayed coating (Metco-2 with a sintered bond coating). The high-temperature superheater or reheater tubes are made of high-chromium steel or austenitic stainless steel. They are left uncoated because the intrinsically formed protective oxide layer on the surfaces has a high resistance to wastage (Wu 2006).

In the early plants (Värtan, Tidd and Escatrón), the furnace membrane walls were left uncoated and subsequently experienced various degrees of wastage. In Värtan, those parts of the membrane wall which were exposed (e.g. tubular offsets for inspection openings) showed erosion damage caused by the return flow of the bed material along the wall (Schemenau and Anderson 1992). However, the membrane walls performed satisfactorily following the application of suitable refractory coatings (Wright et al. 2003).

The material performance of the evaporator tubes depended on the steam conditions and the corrosive environment. At the Värtan plant, one of the two units experienced a failure of the Metco-2 coating on the evaporator tubes. The tube bundle was replaced with a new one, incorporating modifications to the bed flow pattern in order to minimise erosion, and alternative coatings were tested. The Escatrón plant also experienced flaking of the coatings in some local areas. However, the Metco-2 coating performed well at the Tidd plant. There was only minor tube erosion, apparently resulting from local flow disturbances near the bottom of the tube bundle. This might be attributed to the lower steam pressure at Tidd, and thus a lower tube surface temperature than at the Värtan and Escatrón plants (Wright et al. 2003).

Overall, the uncoated superheater tubes performed satisfactorily. The Värtan and Tidd plants experienced some wastage in locations exposed to increased particle flow. This can be treated by taking suitable measures such as heat exchanger redesign or coating (Bauer and Marocco 1995). Only minor erosion damage was detected in the Escatrón plant. The significantly stronger erosion in the Värtan plant can be attributed to the different ash behaviour within it (Jansson 1995a).

### Sintering

Sintering inside the fluidised bed and in the cyclones causes severe problems. In higher temperature zones in the fluidised bed, bed material sinters and forms agglomerates. These are no longer fluidised and stick to heating surfaces and walls. This interferes with the heat transfer of the fluidised bed and impairs steam production. In extreme cases, fluidisation of the bed cannot be maintained. Improvements

to the fuel injection in order to set a homogeneous temperature distribution counteract the sintering processes. Furthermore, uncooled tubes should be avoided in the fluidised bed because these “hot spots” can cause sintered deposits. Such deposits will then accumulate in precisely these “hot spots”. Sintering phenomena also occur in the cyclones: deposits form inside the cyclone, then break off during load changes, clogging the ash discharge from the cyclones (Martinez Crespo 1995).

In Escatrón, both plant components – the fluidised bed and the cyclone – are affected by sintering. As a remedial action, the fluidised bed is operated at a lower temperature and power output. Low fluidised bed temperatures prevent clogging due to sintered pieces in the cyclones. At the plants in Tidd and Värtan, problems also arose with limestone, but not with dolomite. No sintering phenomena were detected at the Wakamatsu plant (Käser 1996).

Sintering depends on the composition of the coal ash. The alkalis in the fuel lower the deformation point of the ash particles, which stick together and sinter. The additives used for desulphurisation have a similar effect. Limestone intensifies sintering more than dolomite. However, the reason behind this has not yet been clarified. One possible reason is that lower deformation and sintering temperatures develop when the mixture features limestone with other ash components, such as alkali compounds, than with dolomite (Käser 1996). Another cause may be the different comminution behaviour during calcination. It is assumed that the pressure, with limestone, suppresses calcination so that larger and heavier particles form. These fail to mix properly in the fluidised bed and are therefore more likely to sinter (Weitzel et al. 1996). The  $\text{SO}_2$  concentration of the flue gas also appears to have an effect. At high  $\text{SO}_2$  emissions and a low desulphurisation degree, the problems which have arisen have been more severe.

The findings collected at the industrial-scale plants in service make it possible to correlate the problems encountered with the alkali contents of the fuel, the  $\text{SO}_2$  content of the flue gas and the additive used. As a result, the suitability of fuels can be assessed in advance (Käser 1996).

### Gas Turbine Erosion

At several plants, erosive wear of the turbine blades was detected (despite the use of erosion/corrosion-resistant coatings) as well as of the inlet guide vanes. This occurred mainly as a result of plugging of the cyclone ash discharge legs. This, in turn, caused an excessive loading of dust and more damaging coarse particulates to be transferred to the turbine. However, when the cyclone ash system performed without problems, the erosive wear was generally limited. At the Escatrón plant, there was lower erosion because of the relatively soft ash which formed from the black lignite used (Wright et al. 2003). Even though the erosion damage does not cause a premature shutdown, it shortens the service life of the machines and raises the maintenance costs. Filtering separators are absolutely essential in order to prevent lifetime-curtailling erosion and fouling of the gas turbines. On the basis of investigations at the Tidd plant, featuring trouble-free operation of the cyclones, a blade lifetime of more than 20,000 h can be expected (Bauer and Marocco 1995).

The lifetimes of turbine blades and in-bed heating surfaces are estimated at 20,000–30,000 h (Renz 1994; Schemenau and van den Bergh 1993).

### **7.3.3.7 Comparison of Bubbling Pressurised Fluidised Beds to Conventional Pulverised Coal Firing**

Pressurisation of the FBC furnace results in a very compact structure, and hence to reduced expenditure on material. A comparison of material requirements for a 330 MW<sub>el</sub> power plant with a pressurised FBC furnace to those with a pulverised fuel fired furnace gives a weight difference of more than 40%. Whereas the equipment requirements for coal feeding and ash removal, mechanical equipment and auxiliary systems in both power plant types are more or less equal, the mass of material for the boiler installation alone is cut by more than 60% for PFBC. Although the weight comparison is not the only tool for comparing the expected costs, it explains the reason behind the development of PFBC furnaces (Rehwinkel 1989).

Based on published data on the Cottbus PFBC plant, the specific costs for a 100 MW<sub>el</sub> plant in condensing operation were about €2,050/kW<sub>el</sub> in 1999. This is comparable to the cost of other power plant technologies for coal. In the lower output range, the PFBC furnace has an efficiency of about 42% and is thus more advantageous than conventional steam cycles, whose efficiency typically falls below 40%. Cost comparisons for higher outputs show that the capital costs for 350 MW and 800 MW<sub>el</sub> PFBC power plants are equal to, or lower than, pulverised coal fuelled power plants with conventional steam conditions (Weitzel et al. 1996; Wauschkuhn 1994).

### **7.3.4 Pressurised Circulating Fluidised Bed Combustion (PCFBC)**

Thanks to its advantages, circulating fluidised bed combustion (CFBC) has become accepted as the preferred option to stationary fluidised bed combustion for atmospheric operation (see Sect. 5.4.2). These advantages also hold true in operation under pressure. In spite of this, pressurised circulating fluidised bed combustion (PCFBC) has not yet reached a similar stage of development. No large-scale plants have been built to date, even though extensive investigations have been carried out and complete plans for industrial power plants with capacities of up to 150 MW<sub>el</sub> have been prepared (Bauer et al. 1994; DOE 2003a). As a result, no operating expertise has been gained in this area.

One essential characteristic of a CFBC furnace is the decoupling of heat release and dissipation (see Fig. 7.11). All the heat is released via the entire furnace. There are no heating surfaces inside the furnace beyond the cooled furnace walls. The heat is dissipated via the furnace walls and an ash fluid bed heat exchanger. The latter cools the ash that was removed in the cyclone.

The necessary temperatures in the fluidised bed are adjusted by recirculating the cooled ash. Depending on the ash recirculation flow, the temperature of the cooled

ash lies between 400 and 600°C. The output of the circulating PFBC is controlled by the fuel flow; the ash recirculation flow controls the temperature in the fluidised bed and the flue gas temperature. Increasing the output also causes an increase in the ash recirculation flowrate.

In bubbling PFBC furnaces, by contrast, heat release and heat dissipation take place in the fluidised bed. Although this makes for compact plant structures, it couples combustion, pollutant formation and heat transfer. Separating the combustion and heat transfer functions from the steam – water cycle in circulating FB combustion offers several advantages (Rehwinkel et al. 1993):

- Without having to take the in-bed heating surfaces into account, the flow velocity can be increased to about 5 m/s in order to generate a high cross-sectional heat release rate and a compact construction body. By contrast, the speed of a stationary FBC is limited to 1 m/s in order to prevent erosion on the in-bed heating surfaces.
- In the ash fluid bed heat exchanger, the fluidisation speed can be chosen independently of the combustion air quantity. Given the small grain size of the circulation ash, low velocities of 0.3 m/s are sufficient for fluidisation. As a result, erosion problems do not arise.
- The flue gas temperature at the outlet of the CPFBC furnace can be kept constant over the whole load. This is because the output is controlled by the fuel mass flow and the temperature by the ash recirculation flow. In the freeboard, it is possible to set flue gas temperatures of 900–1,000°C, even at low loads and low ash recirculation flows, without impairing the desulphurisation process. As a result, the hot gas temperature always lies at 850°C or above. In stationary fluidised bed combustion, the temperature of the fluidised bed is set by changing the bed height. Because of the low fluidisation speed and the lower mass flow, it is not possible to balance the temperature between the bed and freeboard. The in-bed heating surfaces projecting from the bed at part load cause a further decrease in the freeboard temperature of the stationary PFB. At half load, the temperature ranges around 550°C, compared to 850°C at full load. At an inlet temperature of 550°C, the useful output of the gas turbine falls to zero.
- Any influence of the fuel's grain size on the heat transfer in circulating PFBC can be counteracted by modifying the ash recirculation flow. Depending on the reaction kinetic conditions, the plant can be operated with fine or coarse grains. By contrast, a change in grain size influences the heat transfer in stationary FBC. It is impossible to counteract this effect. This results in changes to the steam production from the in-bed heating surfaces and to the flue gas temperature.
- Air staging is one possible method of reducing NO<sub>x</sub> emissions. Using this method, only part of the combustion air is injected into the fluidised bed via the distributor plate. The remaining combustion air is injected further up in the furnace at a later point in time. In stationary PFBC furnaces, in-bed heating surfaces would be damaged not only by erosion but also by increased corrosion in air-deficient zones.



### 7.3.4.1 Bubbling and Circulating Pressurised Fluidised Bed Combustion: Comparison in a Pilot-Scale Plant

A 15 MW<sub>el</sub> pilot-scale power plant, the original design of which included hot gas filters based on ceramic filter candles, was run in stationary operation from 1989 to 1991. Afterwards, it was rebuilt for circulating operation. The tests in circulating operation were performed up until 1992 (Rehwinkel et al. 1993).

The bubbling PFB boiler is shown in Fig. 7.20. The plant comprised wet feeding for coal and limestone and a multistage compressor for supplying the system with combustion air. The PFB firing, the ash removal system and the hot gas filters were located in the pressure vessel. Designed as a bubbling PFB, its equipment included in-bed heating surfaces with a 4 m high bed and a freeboard above it, also 4 m in height. Operated as bubbling FB combustion, the height of the bed had to be lowered at part load in order to reduce the heat transfer to the in-bed heating surfaces. The excess ash was buffered in a bed ash buffer storage facility.

In 1991, the plant was retrofitted for a circulating process (see Fig. 7.21). The pressure vessel then contained the circulating PFB firing with a cyclone downstream

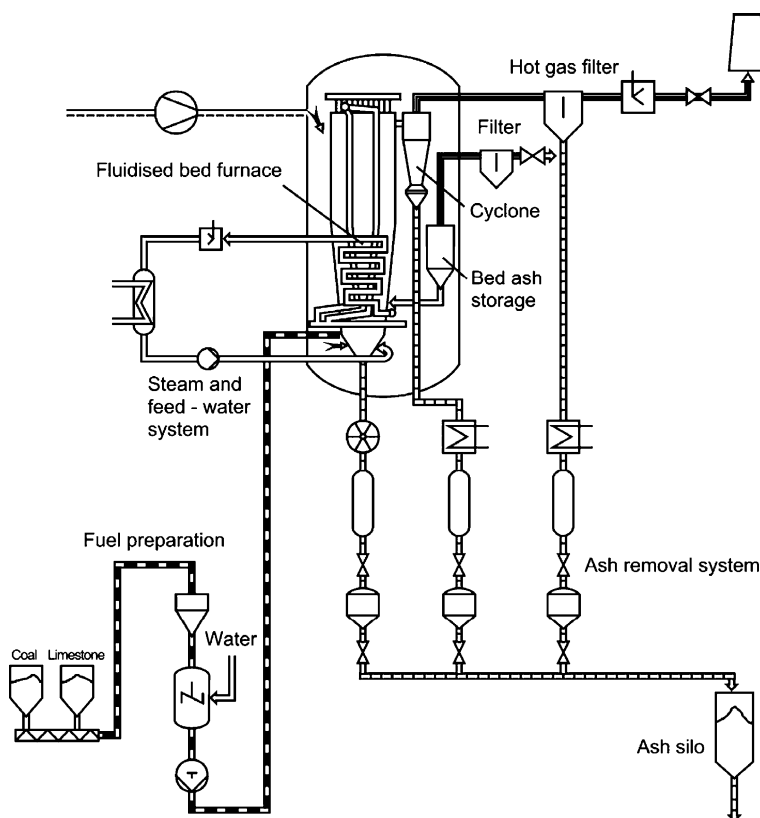


Fig. 7.20 15 MW<sub>th</sub> test plant with bubbling PFB combustion (Rehwinkel et al. 1993)

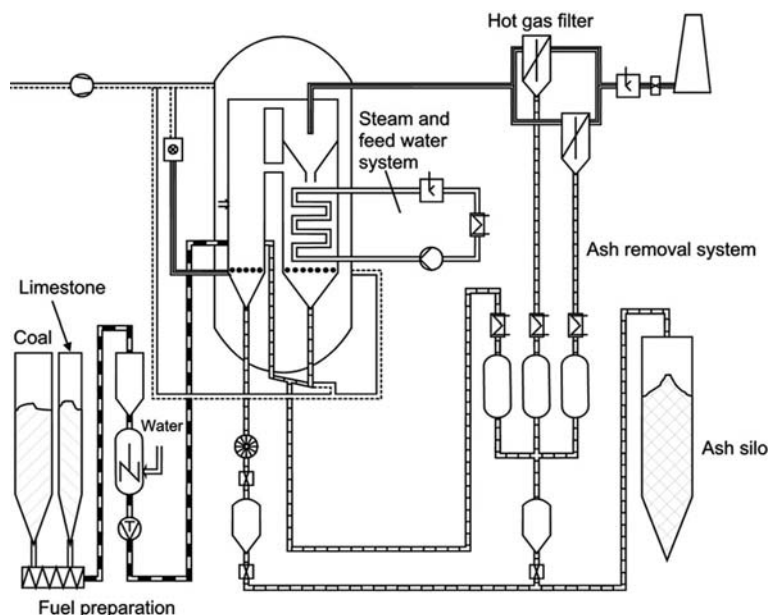


Fig. 7.21 15 MW<sub>th</sub> test plant with circulating PFB combustion (Rehwinkel et al. 1993)

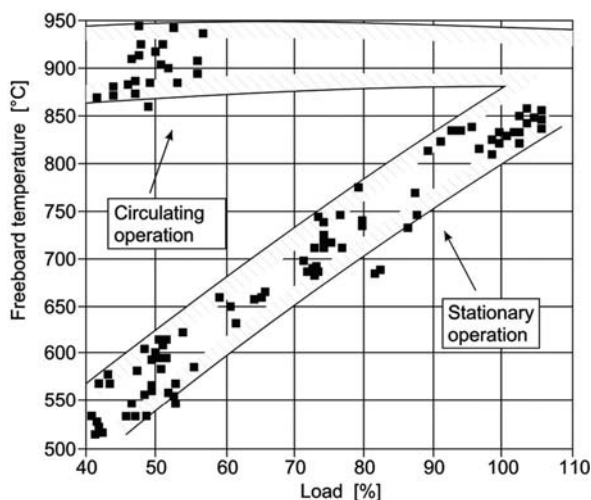
for ash recirculation and a fluid bed heat exchanger to cool the recirculated ash. The bed ash buffer storage facility, which was required for stationary PFB combustion, was removed. The peripheral systems for coal feeding, ash removal, steam cycle and hot gas filtration were left more or less unchanged. The circulating PFB combustion worked with a stationary fluidised bed in the lower part of the combustion reactor to facilitate the admixture of the coal – water suspension. The primary air entered via the distributor plate. Secondary air was injected above the fluidised bed.

The following cyclone removed the ash. In the ash fluid bed heat exchanger, the sensible heat was transferred to the steam – water cycle and directed towards the first pass at 400–600°C. The ash recirculation rate, i.e. the ratio of the ash recirculation flow to the mass flow of fuel and additive fed to the firing, was about 10–20. Lower ash recirculation rates caused the temperature in the freeboard to rise to levels above that of the bed temperatures. By contrast, an increase in the ash recirculation rate caused the excess temperature in the freeboard to fall.

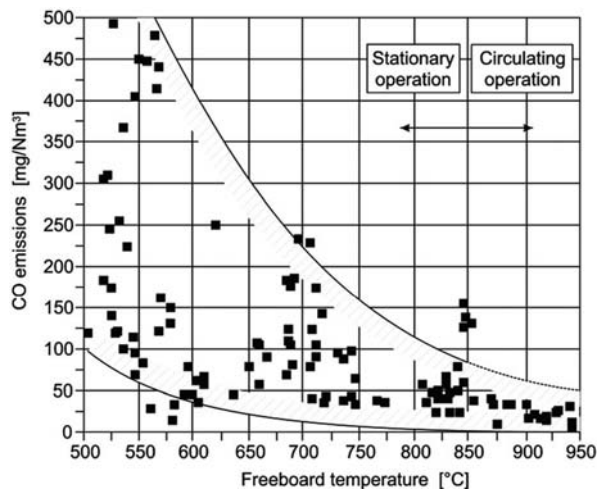
Figure 7.22 shows the freeboard temperatures, measured in tests, as determined by the load. While in bubbling PFB combustion, the in-bed heating surfaces projecting from the fluidised bed at part load caused the freeboard temperatures to fall. These temperatures remained constant in circulating PFB combustion. At full load, the freeboard temperatures in circulating PFB combustion were about 50°C higher than those in the stationary process.

The CO emissions more or less depended on the temperature. The bubbling FBC had low CO emission levels at full load. However, it exhibited a strong increase at part load due to the corresponding decrease in freeboard temperatures. The

**Fig. 7.22** Freeboard temperature as a function of load (Rehwinkel et al. 1993)



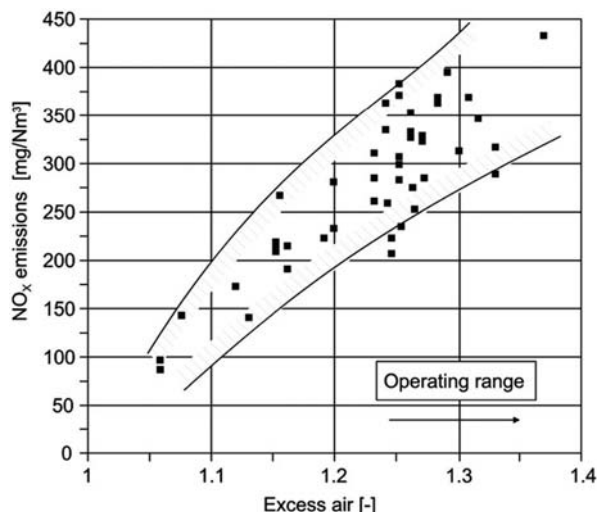
**Fig. 7.23** CO emissions as determined by the freeboard temperature (Rehwinkel et al. 1993)



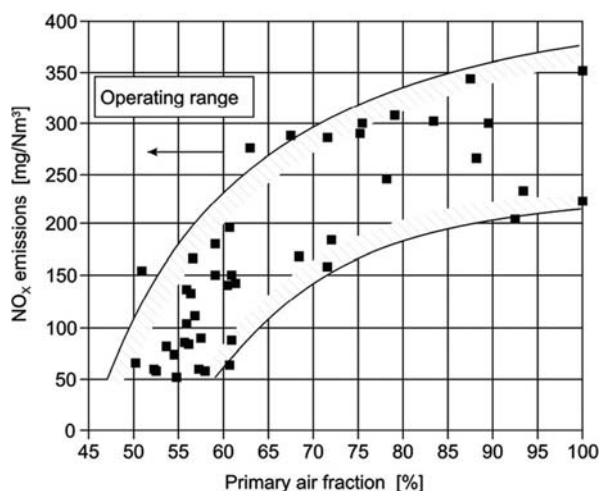
circulating PFBC, at all load ranges, worked at freeboard temperatures equal to, or higher than, the bed temperature. This resulted in low CO emissions. Figure 7.23 shows the CO emissions as determined by the range of freeboard temperatures: below 850°C in stationary and between 850 and 950°C in circulating PFB combustion.

Figure 7.24 shows the  $\text{NO}_x$  emissions as determined by the air ratio for the stationary process. The crucial influence of the excess air ratio is obvious. Because of the potential corrosion of heating surfaces, an excess air ratio below 1.2 is impossible for bubbling PFB combustion. This results in an increase in  $\text{NO}_x$  emissions, which range between 200 and 400  $\text{mg/m}^3$ . In accordance with German emission limits, this makes secondary reduction measures necessary.

**Fig. 7.24**  $\text{NO}_x$  emissions as a function of excess air, bubbling PFBC (Rehwinkel et al. 1993)



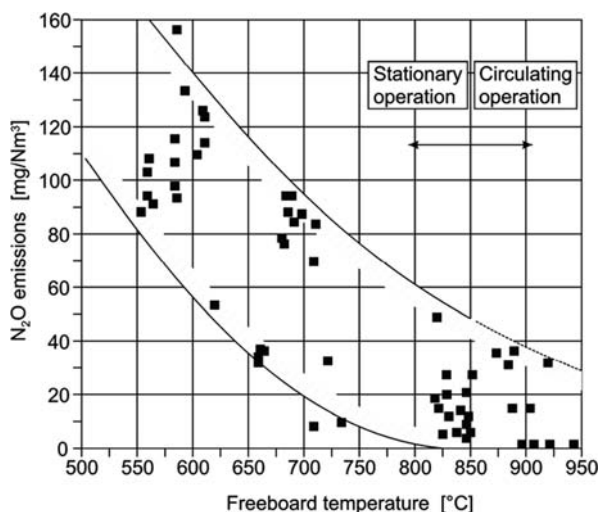
**Fig. 7.25**  $\text{NO}_x$  emissions as determined by the primary air fraction, circulating PFBC (Rehwinkel et al. 1993)



By applying the air staging technique in circulating PFBC – impossible in bubbling PFBC because of the risk of corrosion to in-bed heating surfaces – it is possible to limit  $\text{NO}_x$  emissions without further  $\text{NO}_x$  measures. Figure 7.25 plots the  $\text{NO}_x$  emissions in circulating PFB combustion as determined by the primary air fraction. The operating range lies below a 60% primary air fraction. This produces  $\text{NO}_x$  emissions between 100 and 200  $\text{mg/m}^3$ .

At combustion temperatures below 900°C, typical for bubbling PFB firing, another type of nitrogen oxide develops –  $\text{N}_2\text{O}$  (nitrous oxide, or laughing gas). Tests at a stationary PFBC furnace showed that  $\text{N}_2\text{O}$  emissions are influenced almost exclusively by the temperature. Below 900°C, they rise dramatically as the

**Fig. 7.26**  $\text{N}_2\text{O}$  emissions as determined by the freeboard temperature (Rehwinkel et al. 1993)



temperature decreases. At freeboard temperatures of 850–950 $^{\circ}\text{C}$  in circulating PFB combustion,  $\text{N}_2\text{O}$  emissions can be disregarded. Figure 7.26 shows the  $\text{N}_2\text{O}$  emissions of the bubbling and circulating PFB combustion as determined by the temperature for both cases.

The test results obtained at the pilot-scale plant show how much more advantageous circulating PFB combustion is than bubbling (Rehwinkel et al. 1993). These advantages include

- lower emissions,
- higher part-load efficiency and
- a wider range of fuels.

Studies and plans for circulating PFBC furnaces have been undertaken based on expertise gained with the test plants.

Four companies – VEAG, LLB, Steinmueller and Siemens (Bauer et al. 1994) – planned a power plant with pressurised circulating fluidised bed combustion and a capacity of 150  $\text{MW}_{\text{el}}$ . Raw brown coal was used as the fuel. The planned efficiency of the plant was 45%, with a gas turbine output fraction of 46 MW and a steam turbine power output of 112 MW. The gas turbine inlet temperature was 880 $^{\circ}\text{C}$ , the pressure 16 bar. The steam parameters were 190 bar/580 $^{\circ}\text{C}$ /585 $^{\circ}\text{C}$  (see Fig. 7.27). By awarding the contract for the thermal power plant to Cottbus, the bubbling FBC furnace was chosen over the PCFBC concept.

It was planned to build a 137  $\text{MW}_{\text{el}}$  PCFBC plant in Lakeland, FL, USA, under the US DOE Clean Coal Technology Demonstration Program. The plant would have used Foster Wheeler's pressurised circulating fluidised bed (PCFB) technology and Siemens Westinghouse's hot gas filtration system. The projected net efficiency was 36% (HHV basis). However, this project was cancelled because technical and economic issues could not be resolved. A subsequent plan to convert the installation

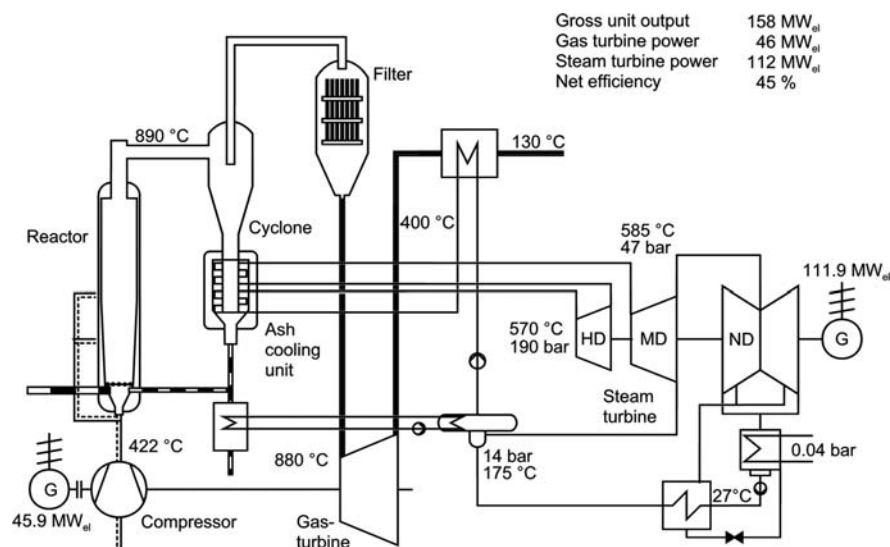


Fig. 7.27 Projected 150 MW pressurised CFBC furnace (Bauer et al. 1994)

into a second-generation PCFBC by adding a carboniser was developed. This is discussed in Sect. 7.3.5 (DOE 2003a; Wu 2006).

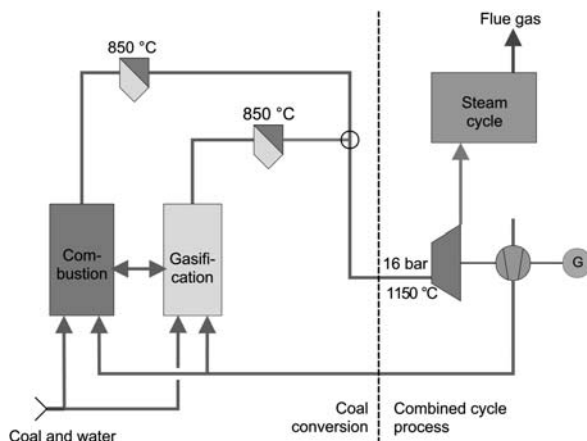
### 7.3.5 Second-Generation Fluidised Bed Firing Systems (Hybrid Process)

The disadvantage of pressurised fluidised bed firing systems is the limited process temperature of the hot gas before it enters the gas turbine (a maximum of about 900°C). Modern gas turbines are capable of operating at higher temperatures, a potential unable to be exploited by the PFB furnace alone.

This potential for an increased efficiency can only be realised when the temperature of the flue gas after the fluidised bed firing and after gas cleaning can be raised to the common gas turbine inlet temperature by firing a “gas turbine compatible” fuel. The usable gaseous fuel can be either an ash-free fuel such as natural gas or a fuel gas produced by coal gasification (see Fig. 7.28). Such procedures to increase the gas turbine inlet temperature are called hybrid or second-generation pressurised fluidised bed combustion processes (“2nd PFBC”).

It should be noted that higher gas turbine inlet temperatures also present higher purity requirements for the hot gas resulting from PFB combustion. The particulates collection technique currently used is that of two-stage cyclones. This is insufficient for second-generation PFBC processes, as an increase in the temperature would cause the remaining ash particles to soften, resulting in fouling deposits in the turbine. The increased fraction of fines with its enrichment of alkalis causes the

**Fig. 7.28** Schematic of a second-generation PFBC



deformation temperature to fall to about 1,000–1,050°C, which is much lower than the deformation temperature of the original coal ash of between 1,150 and 1,250°C. As a result, a hybrid scheme requires a more effective particulate collection system, as well as a hot gas cleaning stage which extracts gaseous alkalis by means of getters. Otherwise, the purity requirements for the gas turbine cannot be met (Jansson 1996).

In the PFBC furnaces currently in use, the output of the gas turbine is one fifth of the overall output. This low figure is due to the cooling of the furnace by steam and water. As a result, an increase in the gas turbine inlet temperature does not cause a corresponding increase in efficiency. A higher efficiency is achieved if the entire fuel is fed to the gas turbine and steam is produced only in the heat recovery boiler. In this case, either the fluidised bed needs to be cooled by a higher excess of air or the heating surfaces have to be air-cooled. The additional gaseous fuels react with the excess oxygen of the hot gas in a special gas turbine combustion chamber. This causes the hot gas temperature to increase to the permissible gas turbine inlet temperature. In this configuration, the output of the gas turbine is about 50–60% of the total output (Rehwinkel et al. 1993). However, cooling the fluidised bed by means of excess air or air-cooled heat exchangers is not state of the art. Those second-generation PFBC systems featuring steam cooling of the fluidised bed combustor which are currently under consideration have a lower share from the gas turbine of the total output. Therefore, the potential for efficiency increases is smaller than for the competing coal-based combined cycles (IGCC, PPCC, EFCC).

Figure 7.29 shows second-generation pressurised fluidised bed combustion as proposed and developed by Foster Wheeler. Coal is fed into an air-blown pressurised bubbling fluidised bed partial gasifier, called a carboniser. The air is supplied in sub-stoichiometric quantities, and the carboniser operates at 900–950°C and 19 bar to produce a low calorific, coal-derived syngas and a char-sorbent residue. After passing through a cyclone and a ceramic barrier filter to remove particulates and alkali vapours, the hot syngas is burned in a specially designed gas turbine

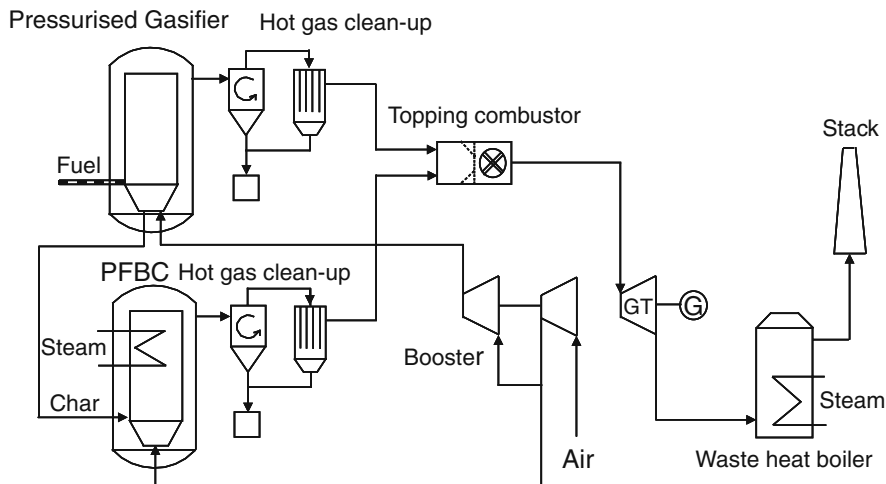


Fig. 7.29 Foster Wheeler's second-generation PFBC concept (Nagel 2002)

combustor called a topping combustor. The gas turbine drives a generator, and its compressor feeds air into the carboniser and the PCFB boiler. The carboniser char is burned in the PCFB boiler using a high amount of excess air to produce superheat and reheat steam for the steam turbine. The hot, oxygen-rich PCFB boiler flue gas passes through its own cyclones and ceramic barrier filters to facilitate the removal of particulate and alkali vapours and the combustion of the syngas in the topping combustor. The efficiency of the design case is 46% (HHV basis); both filters have a temperature of 870°C, the gas turbine firing has a temperature of 1,480°C and the steam is 160 bar and 565°C/565°C. Reducing the temperature of the syngas filter to 540°C and the PFBC filter to 650°C decreases the efficiency to 45.8%. Using double reheating and advanced steam conditions of 275 bar/595°C/595°C/595°C increases the efficiency to 50.5% (Robertson et al. 2005; Robertson et al. 2001).

A second-generation fluidised bed power plant was developed by Foster Wheeler at a pilot-scale facility to test the carboniser, the PCFB boiler, candle filters and the topping combustor. The plant had a coal feed rate of 2,500 kg/h. The tests showed that the original 870°C design temperature of the filter behind the PCFB was not feasible. The limit was determined more by the properties of the ash than by those of the candles. The operating limit was 760°C for bituminous coal, but was about 540°C for petroleum and sub-bituminous coal. Operating at lower temperatures would also mean that an alkali getter would not be required (Wheeldon et al. 2001).

A large-scale demonstration of Foster Wheeler's advanced PCFBC was originally planned in Lakeland, FL, USA, under the US DOE's Clean Coal Technology Demonstration Programme. The plant would have had an output of 240 MW<sub>el</sub>, a gas-to-steam turbine output ratio of 28% and a net efficiency of 40.6% (HHV basis). A net efficiency of 45% (HHV basis) was indicated for greenfield operation. The design temperature of the gasifier was 950°C, while that of the syngas filter was 650°C. The gas turbine firing temperature was 1,290°C. However, it became clear



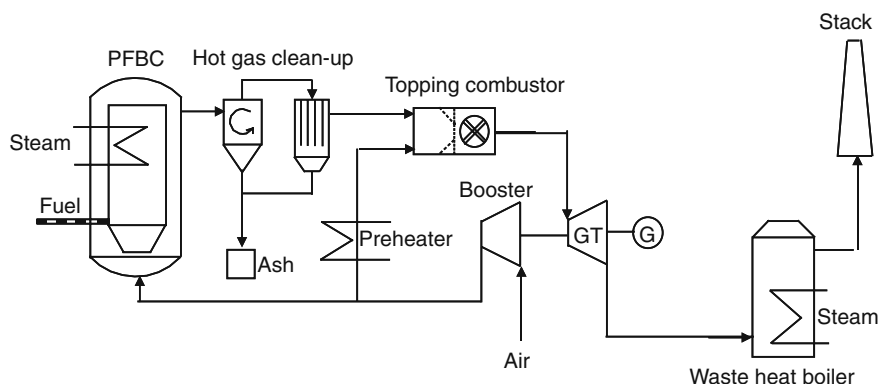


Fig. 7.30 Schematic of a pressurised fluidised bed with staged combustion (Nagel 2002)

that no suitable gas turbine was available. As a result, a decision was taken towards late 2003 to terminate this project (DOE 2003b).

Another possible method of implementing a hybrid scheme is air-deficient operation of the fluidised bed firing followed by flue gas cleaning at the fluidised bed temperature. The addition of air causes the conversion of the unburned gas components and an increase in temperature (Moersch et al. 1999; Nagel et al. 1998).

Figure 7.30 shows a pressurised fluidised bed with staged combustion. Combustion takes place at sub-stoichiometric conditions with air ratios of 0.6–0.8 and temperatures of 800–900°C. Before the particulates are removed by the cyclone and ceramic candle filters, the temperature is lowered to below 700°C. The LCV gas (low calorific value gas with an LHV much less than 10 MJ/m<sup>3</sup>) produced in the first stage is then combusted in a separate combustion chamber to increase the temperature to 1,200°C prior to expansion in the gas turbine. The major advantage of this process over the configuration shown in Fig. 7.29 is the considerably lower capital cost, since only one reactor is required (Nagel 2002). The process has been investigated in greater detail at a pilot scale (Chalupnik et al. 2001).

### 7.3.6 Summary

There is little market potential for PBFBC. While the technology has been demonstrated in several parts of the world, hot gas clean-up remains a key issue. PBFBC also appears to offer less scope for efficiency increases and environmental performance improvements than does IGCC. In addition, it is more complex than supercritical PCC and CFBC, which can offer comparable efficiencies. All these factors may have resulted in the current market difficulties. Alstom, the original developer of PBFBC, is no longer actively marketing the technology, although it does maintain support for existing installations. PBFBC is thus “stalled” at present, and its future development and deployment have become uncertain. If this technology is to be

taken forward, it is more likely to be in the form of advanced PBFBC/PCFBC, which offers greater scope for efficiency increases (Wu 2006).

## 7.4 Pressurised Pulverised Coal Combustion (PPCC)

### 7.4.1 Overview

The aim of pressurised pulverised coal combustion (PPCC) is to produce a hot gas at a high temperature and pressure by the direct combustion of pulverised coal and then to convert its energy into electrical energy in a gas turbine. A downstream heat recovery steam generator (HRSG) and steam turbine extract more electrical energy from the hot gas. The principle of PPC combustion is shown in Fig. 7.31. In the furnace, pulverised coal is burned, producing temperatures of 1,400–1,600°C (which is above the ash fluid point) and a pressure of around 18 bar. Upstream of the gas turbine, it is necessary to remove the molten slag and the gaseous alkali compounds from the hot gas. The heat recovery process, after the hot gas has been cleaned, corresponds to a combined cycle process which is fuelled by natural gas or crude oil. Additionally, though, a DeNO<sub>x</sub> and a desulphurisation stage for the flue gases is necessary at the cold end in order to comply with allowed emission limits.

Pressurised pulverised coal combustion, compared to other advanced methods of power generation by coal, has the potential for the highest efficiency. The PPCC process reaches efficiencies of 53% at ISO gas turbine entry temperatures of 1,200°C (see Sect. 7.1 for a definition of the ISO temperature in relation to gas turbines). Further development of gas turbines towards higher turbine entry temperatures are expected to increase the PPCC process efficiency up to 55% (Schuknecht 2003). In

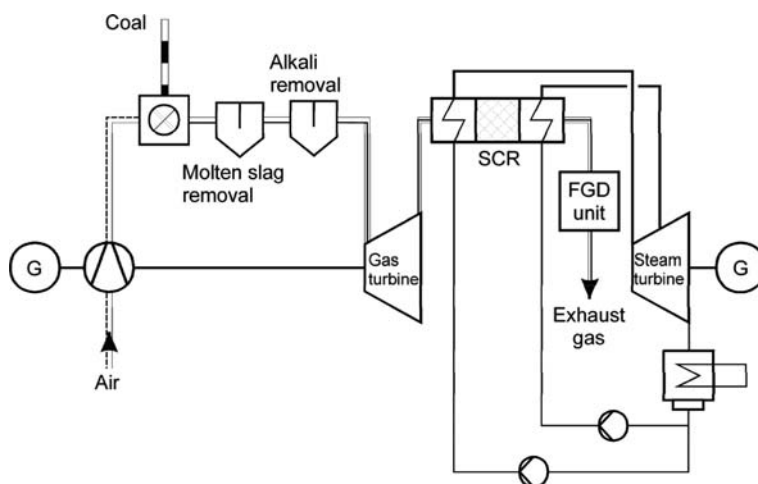


Fig. 7.31 Schematic diagram of a pressurised pulverised coal firing system (Förster et al. 2001)

contrast to technologies that are already demonstrated at an industrial scale (thermal power plants with advanced steam conditions, pressurised fluidised bed combustion and coal gasification), PPCC has not yet reached the demonstration stage. Investigations to date have focussed on slag and alkali removal after the furnace process. Research aiming at implementing the concept of “directly pulverised coal fuelled gas turbines” has been carried out at pilot-scale plants in the USA (Parsons and Byam 1989) and in Germany (Hannes 1996; Hannes 2002; Förster et al. 2005; Förster et al. 2001). Development was discontinued in the USA after approximately 1990, while development in Germany ended in 2005.

All PPCC designs feature in common combustion occurring above the ash fluid temperature, which means part of the ash can be removed early, in its molten state in the furnace. Because the hot flue gas is supposed to serve directly as the working medium of the gas turbine, cleaning of slag particles from the gas is necessary to meet “turbine-compatible” particulates concentration limits. For the conditions of PPCC, a maximum particulate content of  $3 \text{ mg/Nm}^3$ , a maximum particle diameter of  $3 \mu\text{m}$  and a maximum alkali content of  $0.01 \text{ mg/Nm}^3$  STP are desired (Förster et al. 2005).

The designs differ in the gas turbine entry temperature (see Fig. 7.32). The research carried out in the USA focussed on gas turbine entry temperatures of about  $1,000^\circ\text{C}$  (Parsons and Byam 1989). The reduction of nitrogen oxide emissions was achieved by running the combustion in two stages. Cleaning of the flue gas was

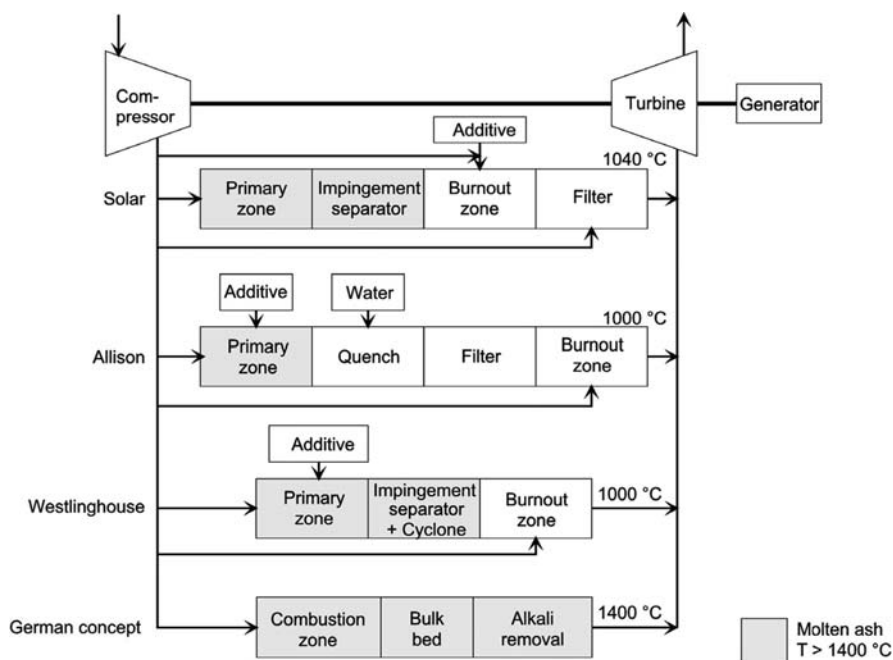


Fig. 7.32 PPCC concepts (Thambimuthu 1993)

carried out either in or immediately after the furnace at high temperatures, or in a range somewhat above the gas turbine entry temperatures, after cooling the flue gas by air or water vapour. The concept favoured in Germany featured a one-stage combustion process and flue gas cleaning at temperatures of 1,400–1,500°C with molten ash removal. The flue gas was conducted to the gas turbine without further cooling (Hannes 1996).

For all PPCC designs, particulates removal at high temperatures and the impacts of gaseous substances forming in the process of combustion at high temperatures are problematic factors. Essentially, these gaseous substances are the alkalis in the flue gas. The alkalis condense in the gas turbine and, in the relevant temperature range, lead to deposition on the turbine blades and to corrosion.

In both the US and the German PPCC designs, hot gas cleaning is at least partly carried out at high temperatures above the ash fluid point. While the German concept uses only high-temperature cleaning, the US concept, with temperatures of about 1,000°C, has the option of cleaning the hot gas both above the ash fluid and below the ash deformation temperature.

Hot gas cleaning, besides slag components, has to remove gaseous gas turbine incompatible components as well. In the following, only the techniques of molten ash removal will be discussed, as the dry ash removal techniques have already been covered in the context of pressurised fluidised bed combustion.

### ***7.4.2 Molten Slag Removal***

Given the high temperatures, molten slag removal is the only option considered for PPCC with high gas turbine entry temperatures. Therefore the gas cleaning temperature must be above the ash fluid temperature, which depends on the coal-specific composition of the ash. The fluid temperatures of hard coal ashes range between 1,350 and 1,500°C. Since thin fluid ash with a low viscosity can be removed more easily, the operating temperature of the gas cleaning should be about 100°C above the fluid temperature. Compared to separation techniques for solid ash particles, the removal of molten ash has only had limited development. Investigations have been carried out at pilot-scale plants in Germany and the USA within the framework of developments of PPCC furnaces (Thambimuthu 1993; Weber and Pavone 1990; Weber et al. 1993; Hannes 1996; Hübner et al. 1988; Förster et al. 2001; Hannes 2002; Förster et al. 2005).

Filtration separators using ceramic filter candles have a high efficiency in the temperature range of that after a fluidised bed combustion zone. For the higher gas temperatures and the conditions of PPCC, suitable filter materials are not yet available. Ceramic filters which have as a base aluminium silicate, aluminium oxide and silicon carbide are attacked and destroyed by molten slag (Hübner et al. 1988). Furthermore, it is unclear whether the removal mechanism of ceramic filter candles, based on surface filtration, can also be applied to liquid matter or molten ash. On the one hand, the risk is that molten ash may penetrate through the pores; on the other,

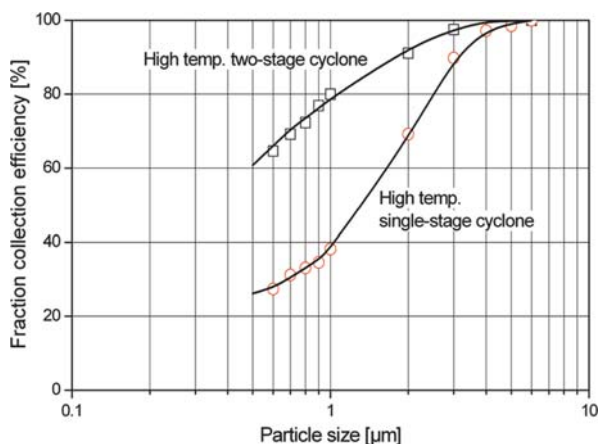
molten ash may hinder the gas penetration when small pore cross-sections are used. The filtering effect that the layer of dust deposition has in the lower temperatures of FBC processes does not occur for PPCC (Weber et al. 1993).

Separation by ESP in the temperature range of PPCC is not possible because the conductivity at such temperatures means a corona cannot form on the spray electrodes (Förster et al. 2001).

Investigations into molten ash removal focus almost exclusively on mass force separators. Among these, inertia and centrifugation separators are the most suitable technologies, since they are much more efficient than gravitation separators. Mass force separators only play a minor role nowadays in conventional dust collection technology, removing solid particles from flue gases under moderate temperature and pressure conditions. Their use has declined because they do not collect small particles and so yield only modest removal rates. The case can be different when molten particles have to be removed. They are more likely to merge before or during the removal process into larger, heavier drops and also stick to the wall of the collector, so that the secondary flows interfering with the gas cleaning process are of minor importance (Weber et al. 1993).

Different variants of slag removal were investigated by the project partners of the combined project “Pressurised Pulverised Coal Combustion”. For the most part, results were obtained from tests at atmospheric plants and later verified in part in pressurised operation (Weber et al. 1993).

By means of a centrifugation separator (a two-stage cyclone) it was possible to achieve outlet particulate loadings of the cleaned gas as low as  $20 \text{ mg/Nm}^3$ . Figure 7.33 shows the fraction collection efficiencies of the cyclone separator measured under atmospheric conditions. According to the diagram, the particle sizes are very small. In contrast to solid ash particle removal, molten ash removal is much more efficient despite the higher temperatures and the higher gas viscosity. This can be put down to the collision and agglomeration of small droplets, as mentioned above, and the drops deforming and sticking when they impinge upon the wall.



**Fig. 7.33** Cyclone removal rate in PPCC as a function of particle size (Weber et al. 1993)

Unlike solid particles, they lose impingement energy through deformation. Also, dispersion of the removed particles is impossible because of the liquid consistency.

From the point of view of material engineering, fewer problems are expected from the use of cyclones because they can be refractory-lined. However, industrial-scale plants with pressurised pulverised coal firing would need a great number of cyclones of a small diameter to achieve high removal efficiency rates – which means the configuration and homogeneous flue gas charging of the cyclones could pose a problem (Weber and Pavone 1990).

Another concept investigated involves a venturi scrubber connected to a cyclone separator. In the venturi scrubber, molten slag is added vertically to the speeding flue gas flow (of a velocity of about 100 m/s) in order to agglomerate the molten ash particles. Therefore a coarser particle spectrum is fed into the cyclone, which results in a higher removal efficiency. Tests carried out discontinuously with slag under atmospheric conditions at temperatures up to 1,600°C showed a reduction in the amount of particulates after the cyclone to 7 mg/Nm<sup>3</sup>, as opposed to 32 mg/Nm<sup>3</sup> without a venturi scrubber. The addition of molten slag is assumed to also favour a higher alkali capture in the slag (Hoberg and Gudenau 1998). Problems in applying this concept may arise with liquid ash recirculation, necessary for a continuous process, and the materials' ability to resist corrosion by molten ash.

Although packed-bed or granular bed filters for solid dust particle removal are classified as filtration separators, the removal of liquid matter by such means must be classed as separation by inertia and mass force. Divided into many sub-streams, the untreated gas flows through the packed bed. Each sub-stream is deflected by bulk material, and droplets, due to their inertia, impinge upon it. The packed bed offers sufficiently many flow paths for the slag to drain out. Outlet particulates loadings of the cleaned gas between 50 and 200 mg/Nm<sup>3</sup>, depending on the filter velocity, are reported for atmospheric tests (Hannes 1996).

The developments at the PPCC test facility at Dorsten in its last years focussed on molten slag removal in a packed-bed filter (Förster et al. 2005; Hannes 2002; Förster et al. 2001). As a result of the long years' tests, it was concluded that inertia separators, such as the packed-bed filter, are not capable of achieving concentrations lower than about 350 mg/Nm<sup>3</sup> or removing particles with a diameter smaller than 3 µm. The removal rates of the pressurised 1 MW facility were considerably higher than the previously reported atmospheric small-scale test results. Tests with higher velocities, increased bed heights and smaller bed material did not improve the removal efficiency.

This is explained by the electrical behaviour of the flue gas, which can be considered to be a cold plasma at the existing process temperatures, meaning that the flue gas behaves neutrally macroscopically, but a separation due to electrical charges occurs. This fact can be verified by the conductivity of the flue gas, as well as by the charges of the particles. The positive charges of the small particles cause an electrostatic repulsion, so that small particles can no longer agglomerate and therefore cannot be separated.

This knowledge provided the basis for developing a novel concept for fine particulates separation by exploiting the electrical properties. By disassociating the

charges from the particles, the resulting non-charged particles can agglomerate and be separated by adhesion on the bed material. Employing a suitable electrical field can enhance the separation of charged particles. Disassociating the charges requires conductive ceramic materials such as those containing  $\text{ZrO}_2$ . Investigations were carried out to design a local electrical gradient by combining suitable ceramic materials and employing SiC (silicon carbide) electrodes behind the packed bed. As a result of this development, particulates concentrations below  $1 \text{ mg/Nm}^3$  with all particles smaller than  $1 \mu\text{m}$  were claimed to be achieved, which would fulfil the requirements of advanced gas turbines (Förster et al. 2005).

### 7.4.3 Alkali Release and Capture

#### 7.4.3.1 Fundamentals

##### Alkali Release

Coal ashes contain the alkalis sodium (Na) and potassium (K) in very differing concentrations depending on the coal type. Hard coal shows typical concentrations in the order of magnitude of 0.5–6, with a maximum of 15% by weight of the coal ash. Alkalis are organically bound in the coal matrix or exist in the mineral phase either as simple salts or as complex aluminosilicates. The type of bond has a major impact on the behaviour of alkalis during the combustion process.

Alkalis are released from combustion as gaseous components mainly in the form of NaCl and KCl or else retained firmly bound in the coal ash. Alkalis released during combustion are termed “active alkalis” (see Table 7.8). Active alkalis consist primarily of simple, inorganic salts and organically bound alkalis. Those alkalis bound in clay minerals as aluminosilicates stay almost inert during combustion. While for hard coals, alkalis are more frequently bound in the mineral components, younger coals, such as brown coal, have a higher alkali fraction in the organic components (Singer 1991).

The main parameter of the combustion process influencing the degree of gaseous alkali release is the temperature. The alkali release of a fuel is often correlated with the chlorine content of the coal. This correlation is valid for coal types which contain alkalis in the form of simple salts, such as German hard coals. Although not generally applicable to fuels with organically bound alkalis, it has been observed for German brown coals as well (Oleschko and Muller 2007).

Another possible method to determine the active alkali content is by solution in water or weak acids. Depending on the type of solvent used, this method either

**Table 7.8** Classification of alkalis in coal

Active	Inactive
<ul style="list-style-type: none"> <li>• Simple inorganic salts               <ul style="list-style-type: none"> <li>◦ NaCl, <math>\text{Na}_2\text{SO}_4</math>, <math>\text{Na}_2\text{CO}_3</math></li> <li>◦ KCl, <math>\text{K}_2\text{SO}_4</math>, <math>\text{K}_2\text{CO}_3</math></li> </ul> </li> <li>• Organically bonded alkalis</li> </ul>	Clay and schist minerals: Complex aluminium silicates $\text{Na}_2\text{Al}_2\text{Si}_6\text{O}_{16}$ $\text{K}_2\text{Al}_2\text{Si}_6\text{O}_{16}$

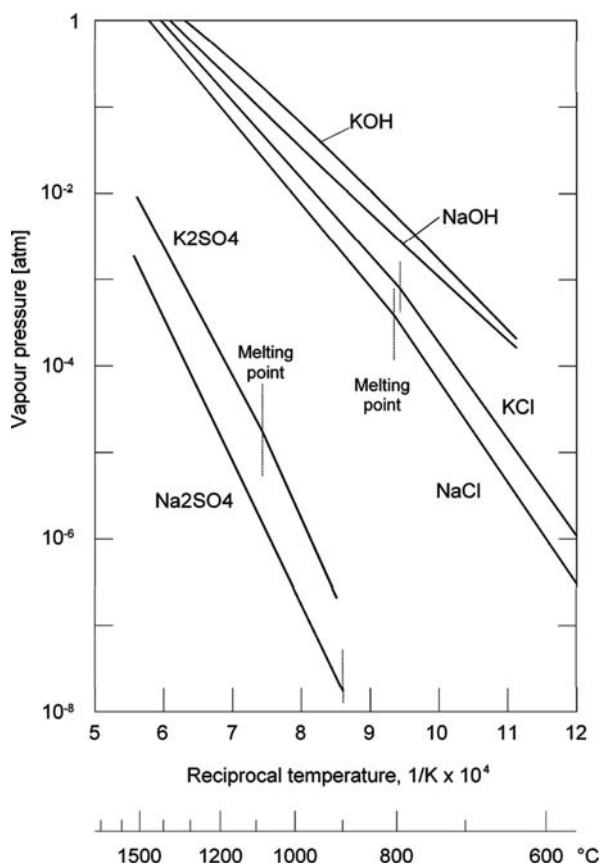
detects only the simple salts dissociated in the coal moisture or, depending on the strength of the acid, organic compounds as well.

Active alkalis contribute substantially to fouling and slagging in conventional firing systems. In combined cycle processes with coal combustion, gaseous alkali compounds condense during expansion in the gas turbine and can, like aerosols, deposit on the blades, inducing fouling and corrosion.

### Alkali Compounds in the Flue Gas and Their State of Aggregation

For a combined cycle process with coal combustion, the type of alkali compounds in the flue gas and their state of aggregation are also of interest, not just the quantity of released alkalis. Alkalis, after release and conversion, are found mainly as low molecular weight compounds such as sulphates ( $\text{Na}_2\text{SO}_4$ ,  $\text{K}_2\text{SO}_4$ ), hydroxides ( $\text{NaOH}$ ,  $\text{KOH}$ ) or chlorides ( $\text{NaCl}$ ,  $\text{KCl}$ ).

To determine the states of aggregation of the compounds, one way is to employ the vapour pressure, equilibrium states of which are shown in Fig. 7.34 as a function



**Fig. 7.34** Vapour pressures of the chlorides, hydroxides and sulphates of sodium and potassium (Scandrett and Clift 1984)



**Table 7.9** Saturation-phase pressures and concentrations of alkali compounds at 1,173 K (Scandrett and Clift 1984)

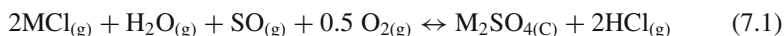
Component	Saturated-state pressures [bar]	Concentration [ppmw]	
		Salt	Na or K
Na <sub>2</sub> SO <sub>4</sub>	$2.94 \times 10^{-8}$	0.014	0.004
K <sub>2</sub> SO <sub>4</sub>	$2.33 \times 10^{-7}$	0.13	0.06
NaCl	$2.12 \times 10^{-3}$	410	160
KCl	$4.86 \times 10^{-3}$	1,200	620
NaOH	$13.68 \times 10^{-3}$	1,800	1,000
KOH	$23.3 \times 10^{-3}$	4,300	3,000

of the temperature. Table 7.9 gives the corresponding saturation concentrations for the saturated-state pressures at a temperature of 900°C.

For chlorides and hydroxides, the saturation concentrations are on a level far above those usually measured in flue gas; for sulphates, they are below. Thus it can be concluded that alkali compounds in the flue gas present as chlorides and hydroxides are found in the form of vapours, and alkali compounds in the flue gas present as sulphates are found in the form of condensates (Scandrett and Clift 1984).

It is assumed that, in the process of the release of alkalis, chlorides and hydroxides form initially and can then be converted by gas-phase reactions (depending on the composition of the flue gas). At the same time, gaseous alkalis can undergo heterogeneous reactions with solids or liquids, which offers the possibility of capture of the gaseous alkalis.

Of particular importance to the alkali speciation is the reaction with sulphur oxides (SO<sub>x</sub>) to form sulphates, according to the following mechanism:



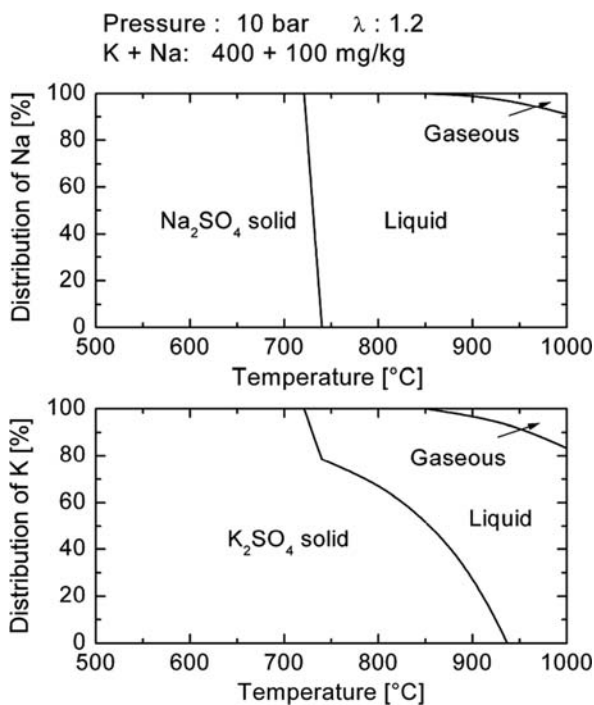
(g) = gas phase, (c) = condensate.

At the high temperatures of combustion processes, sulphur oxides exist as sulphur dioxides, SO<sub>2</sub>. The equilibrium of the alkali reactions with SO<sub>2</sub> depends on the temperature, pressure and flue gas composition. Figure 7.35 gives the results of thermodynamic equilibrium calculations for peat based on the minimisation of the Gibbs free enthalpy (Mojtahedi and Backman 1989).

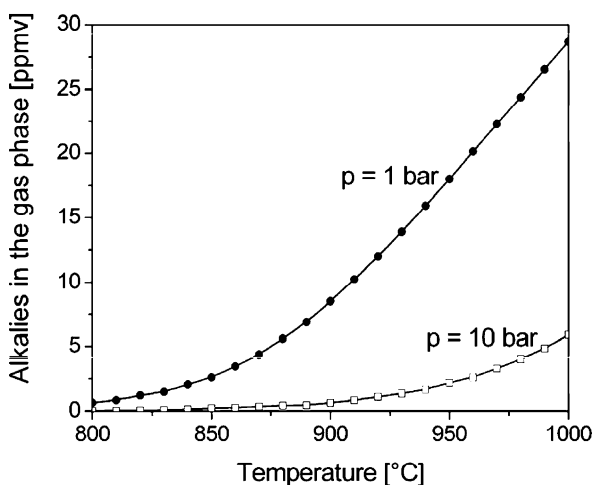
Given fluidised bed temperatures, oxidising conditions and low HCl concentrations, a large fraction of the alkali chlorides react with sulphur oxides to form sulphates, which either condense on particles or in turn form aerosols. The alkalis condensed on particles can be removed by particulate filters. Removal of the condensed sulphate aerosols is more difficult due to their small size (smaller than 1 µm). Filter separators with a deep-layer effect, such as packed-bed filters, seem to be most suitable for this purpose.

The influence of pressure on the gaseous alkalis for fluidised bed temperatures is shown in Fig. 7.36. Higher pressures favour sulphate formation and impede the formation of gaseous compounds. Increasing concentrations of HCl in the flue gas

**Fig. 7.35** States of aggregation of sodium (Na) and potassium (K) compounds under pressurised fluidised bed conditions (Mojtahedi and Backman 1989)

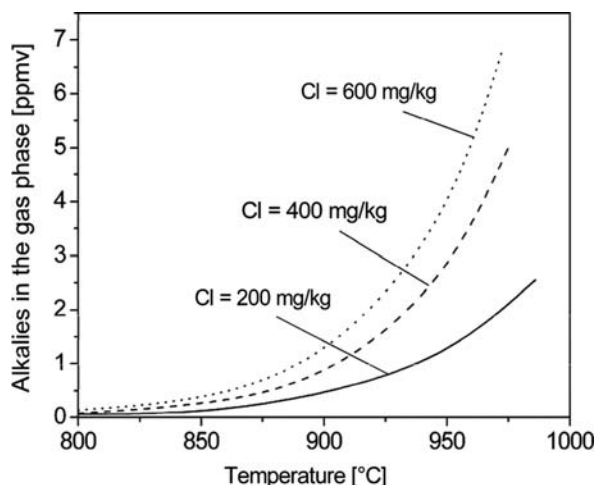


**Fig. 7.36** Effect of pressure on alkalis in the gas phase, data from Mojtahedi and Backman (1989)



shift the equilibrium of the reaction towards the gaseous compounds and impede the formation of sulphates (see Fig. 7.37). The alkali content of the fuel itself only plays a minor role in determining the concentration of gaseous compounds (Mojtahedi and Backman 1989).

**Fig. 7.37** Effect of chlorine content on concentrations of gaseous alkalis, data from Mojtahedi and Backman (1989)



At the higher temperatures of pressurised pulverised coal firing, gaseous chlorides and hydroxides predominate. Elemental gaseous alkalis occur only in trace quantities across the entire temperature range (Hannes 1986).

In conditions without  $\text{SO}_2$ , for example in gasification, no sulphates but only gaseous alkali compounds form, such as chlorides and hydroxides. Two or three orders of magnitude higher levels of gaseous alkali compounds are thus reported for pressurised fluidised bed gasification in contrast to pressurised fluidised bed combustion.

### Alkali Removal

Alkalis can be separated by chemical reaction or removed by physical adsorption on solid or liquid sorbents (known as getters). In the case of chemical sorption, the getters become exhausted, while in the case of physical adsorption, they can be regenerated. In the temperature range of fluidised bed combustion, the basic procedure is physical adsorption, possibly followed by chemical capture. Because of its negative temperature gradient, adsorption is not suitable for higher temperatures, which means chemical capture has to be applied.

Suitable alkali getter materials are quartz ( $\text{SiO}_2$ ), aluminosilicates ( $\text{Al}_2\text{O}_3 \cdot \text{SiO}_2$ ), clay ( $\text{Al}_2\text{O}_3$ ) and naturally occurring substances such as bauxite, kaolinite, emathlite or diatomaceous earth, essentially composed of aluminium oxide and silicon oxide (Thambimuthu 1993; Punjak et al. 1989). Table 7.10 shows their composition by weight.

Thermodynamic equilibrium calculations of systems with different getter materials can be used to evaluate their potential for the chemical capture of alkalis. Even though these evaluations are valid only for the state of equilibrium, they are effective to some extent in predicting the practicability of various getter materials.

In principle, a differentiation has to be made between the type of bonding process of the sulphates and the chlorides. In the temperature range of fluidised beds, the

**Table 7.10** Composition by weight of additives for alkali capture (Punjak et al. 1989)

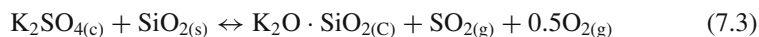
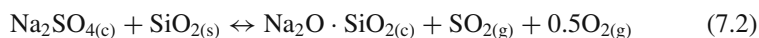
Component	Bauxite	Diatomaceous earth	Kaolinite	Emathlite
SiO <sub>2</sub>	0.11	0.92	0.521	0.734
Al <sub>2</sub> O <sub>3</sub>	0.842	0.05	0.449	0.139
Fe <sub>2</sub> O <sub>3</sub>	0.048		0.008	0.034
TiO <sub>2</sub>	—		0.022	0.004
CaO	—		—	0.05
MgO	—		—	0.026
K <sub>2</sub> O	—		—	0.012
Na <sub>2</sub> O	—		—	0.001

alkali compounds are bound mainly as sulphates, but also as chlorides; at the high temperatures of pressurised pulverised coal firing, binding as chlorides dominates:

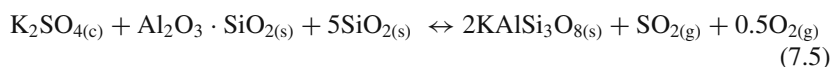
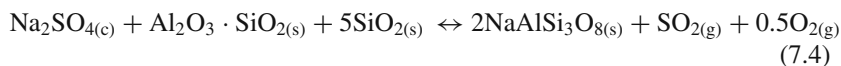
(a) Reactions of the alkali sulphates (Scandrett and Clift 1984):

The chemical bonding of the alkali sulphates can occur according to the following reaction mechanisms:

(1) Silica sand:



(2) Aluminium silicate:

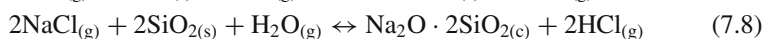
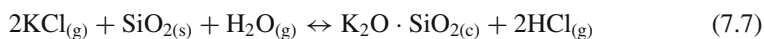
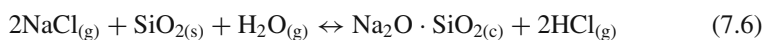


(g) = gas phase, (c) = condensate, (s) = solid matter.

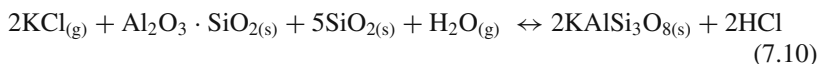
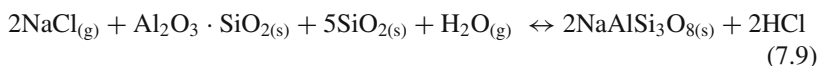
All bonding reactions of alkali sulphates are strongly endothermic reactions and so require a certain temperature to proceed. Reactions with silica sand need a temperature of above 1,300 K to achieve bonding of the sulphates. For a chemical capture process following fluidised bed combustion, reactions with silica sand therefore do not seem suitable at these temperatures. Aluminium silicates react and bond with alkalis at temperatures above 1,000 K.

(b) Reactions of the alkali chlorides (Scandrett and Clift 1984):

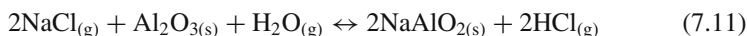
(1) Silica sand:



## (2) Aluminium silicates:



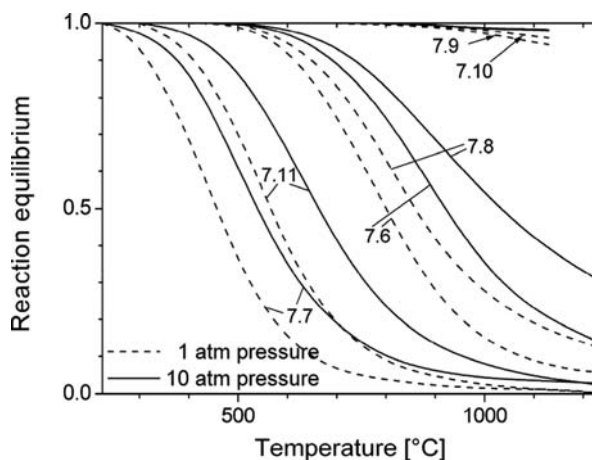
## (3) Alumina:



(g) = gas phase, (c) = condensate, (s) = solid matter.

All bonding reactions of alkali chlorides are strongly exothermic reactions, so the capture rates of all of the getters decrease with rising temperatures. Silica sand getters yield very low capture rates. With pure alumina as a getter, substantial conversion takes place only at temperatures below 800 K (530°C). The use of aluminium silicates, however, makes it possible to achieve a capture rate of 99% at 1,000 K (730°C) and above 95% at 1,200 K (930°C). From this it is evident that aluminium silicates are the only practical material for getters for gaseous alkalis (see Fig. 7.38). For the calculations with pressures of 1 and 10 bar, it was assumed that HCl develops exclusively by reaction and that the water vapour content was 3% by volume. Higher water vapour contents favour, while higher HCl concentrations in the flue gas impede the chemical conversion (Scandrett and Clift 1984).

Thermodynamic calculations substantiate that capturing is also possible at a high-temperature range of around 1,400 °C. At such temperatures, the aluminium silicates mentioned before have an optimal capture rate at an  $\text{Al}_2\text{O}_3$ -to- $\text{SiO}_2$  ratio of 1:8. The alkalis are captured in the liquid phase (Willenborg et al. 2006).



**Fig. 7.38** Equilibrium of alkali capture reactions (Scandrett and Clift 1984)

### Measurements of Gaseous Alkali Compounds

When comparing concentrations of gaseous alkalis, attention must be paid to the measurement method as well as to the measurement location. The ability of each of the measurement methods to detect condensed aerosol particles as well as gaseous components, or only gaseous components, must be known. If the measurement method detects the sum of all the gaseous, liquid and fine particles portion of the solid alkalis, the filter system has the task of separating the liquid and solid alkalis to exclusively feed the sampling setup with gaseous alkalis. While the filtration of condensed and particle-bound sulphates is possible, it is more difficult to remove aerosols, and hence errors of measurement can occur. In the worst case, when the filtration is insufficient, the solid alkali compounds present in small particles are also measured.

Measurements of the gaseous alkalis under fluidised bed conditions are usually carried out behind filters, so that most of the sulphates have already been separated and only gaseous compounds are detected. At higher temperatures, gaseous chlorides and hydroxides predominate instead of liquid sulphates. Measurements obtained at combustion plants are influenced by the alkali release and gas-phase and heterogeneous capture reactions with bed, fly and liquid ash.

Concentrations are given either per unit mass in mg alkalis/kg flue gas or per unit volume in ppmv.<sup>1,2</sup> Wet-chemical measurement methods detect the ions in the solvent, while on-line measurement methods detect alkali atoms or alkali ions in the gas (Bonn 1996). Results of on-line alkali concentration measurements using simultaneous surface ionisation (SI) and excimer laser-induced fragmentation fluorescence (ELIF) have been compared for a PFBC (Monkhouse et al. 2003). Both methods measure in real time but are complementary in that ELIF can discriminate towards gas-phase alkali species, whereas SI detects alkali both in the gas phase and on aerosol particles (Monkhouse 2002).

Mass concentrations can be given either as pure alkali metals or as compounds. For a comparison and the conversion of concentrations per unit mass and per unit volume, it is necessary to know the compound used as a basis as well as the flue gas composition. Usually, however, the relevant data cannot be found in the literature.

By way of example: for alkali chlorides at an air ratio of 1.3, the following conversion holds true:

$$1 \text{ ppmv Na} = 1 \text{ ppmv NaCl} = \frac{1}{2} \text{Na}_2\text{SO}_4 \text{ ppmv} \\ \sim 0.75 \text{ mg Na/kg}(0.75 \text{ ppmw}) \sim 1.9 \text{ mg NaCl/kg} \sim 4.6 \text{ mg Na}_2\text{SO}_4/\text{kg} \quad (7.12)$$

$$1 \text{ ppmv K} = 1 \text{ ppmv KCl} = \frac{1}{2} \text{K}_2\text{SO}_4 \text{ ppmv} \\ \sim 1.27 \text{ mg K/kg}(= 1.27 \text{ ppmw}) \sim 2.42 \text{ mg KCl/kg} \sim 5.65 \text{ mg K}_2\text{SO}_4/\text{kg} \quad (7.13)$$

<sup>1</sup> ppmv = volume parts per million

<sup>2</sup> ppmw = mass parts per million

### 7.4.3.2 Alkali Emissions from Combustion

Most published research into alkali emissions from pressurised fluidised bed combustion has been carried out at a temperature range of about 800–900°C. Only a limited number of measurements have been reported for the conditions of high-temperature pressurised pulverised coal combustion.

Measurements taken after a filter unit under fluidised bed conditions reveal a great scattering of values, from several  $\mu\text{g/kg}$  up to some  $\text{mg/kg}$ . These differences can be put down to the different fuels and test parameters, in particular the temperature. The measurement technology for gaseous alkalis also contributes significantly to this degree of scattering.

To give an example: for peat, a young fuel type, reports give flue gas sodium concentrations between 0.2 and 1.5  $\text{mg/kg}$  and potassium concentrations between 0.07 and 0.4  $\text{mg/kg}$ . These concentrations increase with rising temperatures and decrease with pressure. They are also dependent on the additive for sulphur retention, with dolomite showing lower concentrations than limestone. These concentrations mean conversion rates of the fuel alkalis of 1–4% for sodium and 0.2–0.5% for potassium. In the case of a hard coal, the flue gas sodium concentrations ranged from 0.01 to 0.06  $\text{mg/kg}$  and the potassium concentrations from 0.02 to 0.07  $\text{mg/kg}$  – even though the sodium and potassium contents of this hard coal were equal to or higher than those of peat. Thus the resulting conversion rates are 0.01–0.05% for the sodium in the fuel and 0.01–0.02% for the fuel potassium (Hippinen et al. 1991).

The consideration of the thermodynamics of the reactions of alkali chlorides and hydroxides in forming sulphates in Sect. 7.4.3.1 (Eq. 7.1) suggests that removal in the temperature conditions of pressurised fluidised bed firing should be complete. Tests, however, did not substantiate this. The assumed cause was the presence of HCl, which impedes the formation of sulphates but favours the formation of gaseous chlorides which cannot be removed in the filter (Thambimuthu 1993).

Measurements by optical laser, which only detects gaseous alkali chlorides and hydroxides, show big differences for different coal types. Measured after a cyclone filter for a brown coal type, the concentrations of sodium in the gas phase amounted to 4–5 ppmv and for potassium to 3.5–4 ppmv, whereas for hard coals, the concentrations of sodium and potassium were 30–70 and 10–30 ppbv, respectively. This variability corresponds to the range of dispersion of other measurements. The addition of kaolin reduced the flue gas concentrations by a factor of 2–3 due to capture by the increased amount of aluminosilicate. The use of the hard coal ash as the bed material for lignite combustion resulted in a decrease in concentrations by one order of magnitude. For both coals, potassium was more effectively captured than sodium (Gottwald et al. 2001).

Research into the in situ capture of alkalis in fluidised bed combustion has been carried out at various experimental plants. At one pressurised fluidised bed test plant, combustion tests using a brown coal determined the influence of different bed materials on the alkali concentrations after a two-stage cyclone. With alumina, which consists of 99% aluminium oxide, a reduction ratio of nearly 90% was achieved. The capturing mechanism is based on reactions of alumina with

the silicon compounds in the coal, converting vaporous alkali chlorides and alkali sulphates into high melting point alkali aluminium silicates. Using silica sand, the capture rate was lower. However, the test results show that, even with alumina as an additive, the limit of 0.024 mg/kg was usually exceeded by one order of magnitude (Radhakrishnan et al. 1986). Further investigations with additions of bauxite and kaolinite at another PFBC plant revealed that both materials are suitable for the capture of alkalis. Although tests with a high-alkali young brown coal and with an older brown coal with sodium added to it achieved capture rates of up to 90%, the concentrations of gaseous sodium compounds were in the order of just below 1 mg/kg – thus significantly higher than the previously mentioned limit (Mann and Ludlow 1997).

At the pressurised fluidised bed demonstration plant in Wakamatsu, alkali concentrations of approximately 0.03 mg/kg were detected in the flue gas after a ceramic filter unit, using different hard coals at fluidised bed temperatures of 800°C (Daijou et al. 1997).

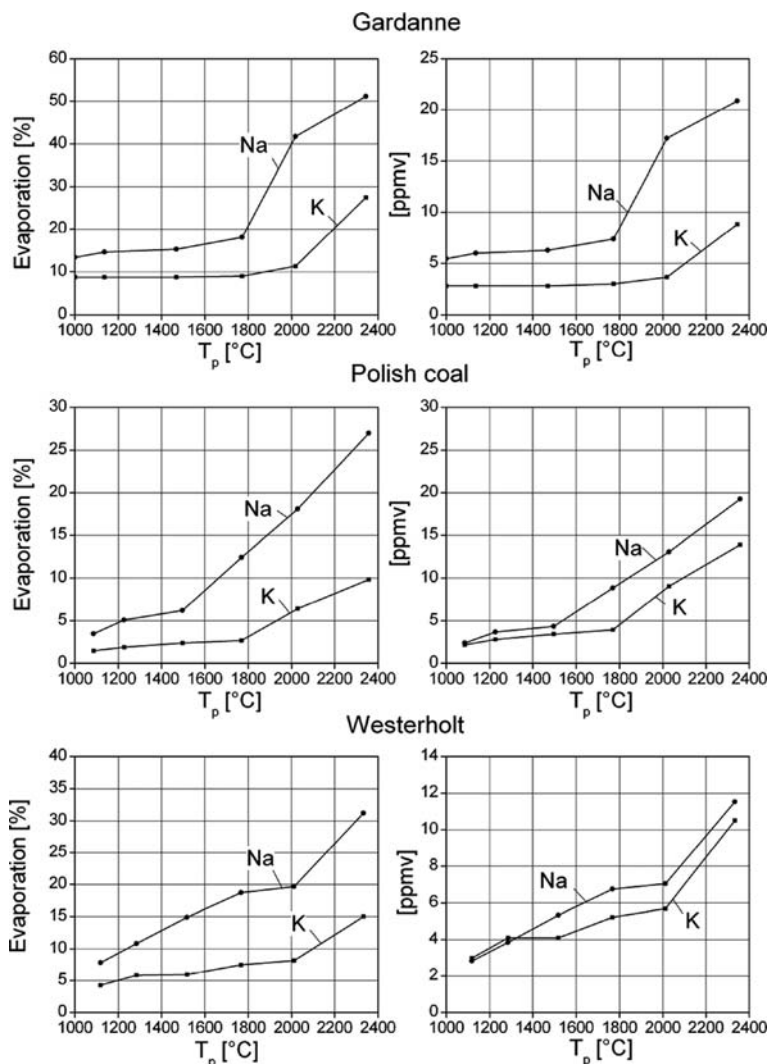
Aho (Aho et al. 1995) carried out systematic tests at an electrically heated entrained-flow reactor at particle temperatures between 1,000 and 2,400°C. The gaseous release of sodium was higher than that of potassium for two hard coals and one lignite. In these tests, the conversion of the sodium was several percent at temperatures somewhat above 1,000°C and between 25 and 50% at higher temperatures (see Fig. 7.39). This data shows that the alkali concentrations, even at low temperatures of around 1,000°C, are at least two orders of magnitude higher than those allowed for gas turbines.

The alkali release was studied during comprehensive investigations by Reichelt at a pressurised entrained-flow reactor (Reichelt 2001). As in the investigations by Aho, the reactor was operated differentially according to the entrained-flow principle. This means that the fuel mass flow is very small in comparison to the volumetric gas flow. The volumetric gas flow, with an oxygen concentration of 6%, is preheated to reactor temperature; the slight loading of the fuel, of 6 g/m<sup>3</sup>, causes minor changes in the concentration in the reactor (i.e. a differential mode of operation). Because of the small fuel load and the corresponding low ash concentration, it is assumed that, in contrast to industrial plants, capturing of alkalis in the solid phase is suppressed to a great extent. For two hard and two brown coals, the gaseous chlorides and hydroxides were detected by means of optical lasers. The results reveal rising alkali emissions with increasing temperatures and falling emissions with increasing pressure. The alkali concentrations presented for a hard coal, at a reactor temperature of 1,400°C, and atmospheric pressure, correlate with a sodium release of about 50% and potassium release of only 20%.

Investigations by other authors indicate a release of sodium of about 50% and of potassium of about 20% for a hard coal at 1,600°C, whereas at temperatures of 1,800–1,900°C, the studies identified an almost complete release (Wen et al. 1992).

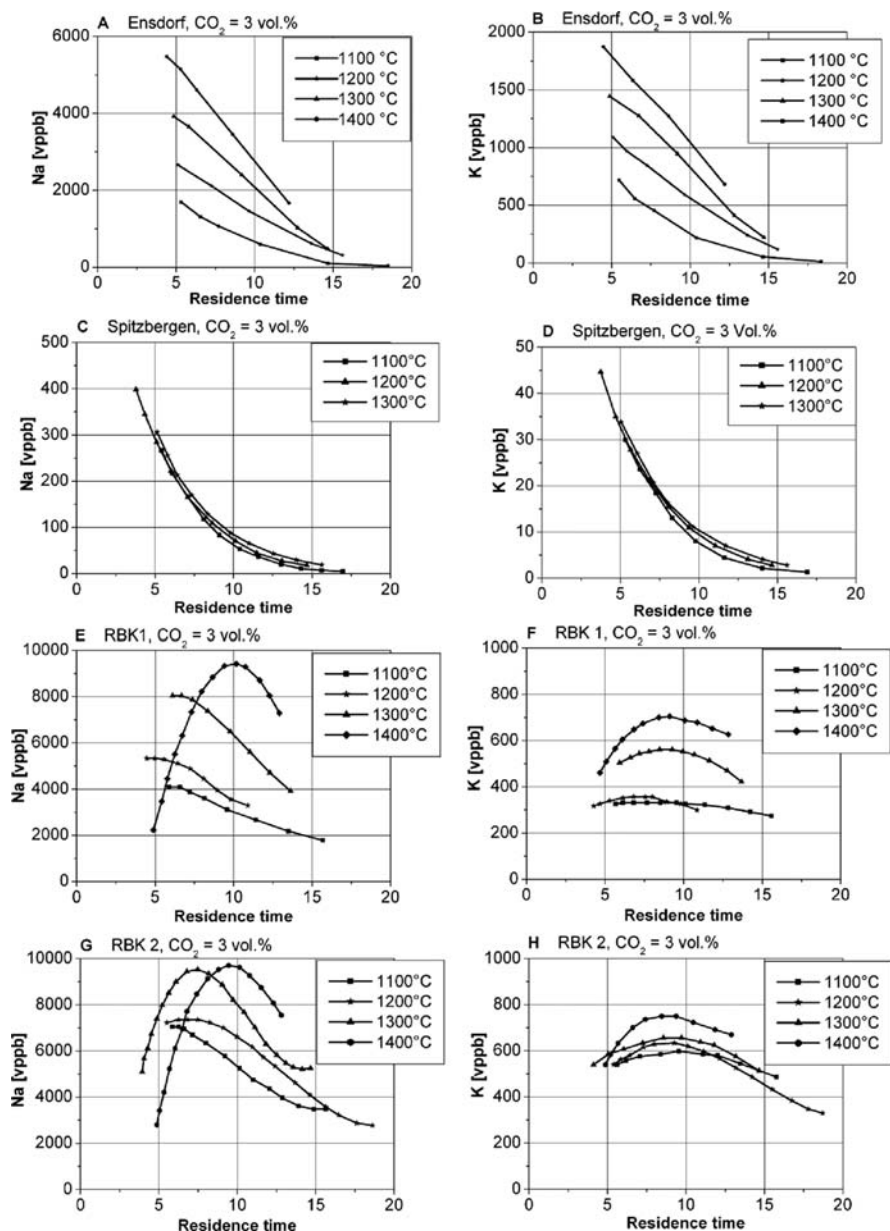
Reichelt (Reichelt 2001) also investigated an atmospherically operated combustion reactor. This reactor was operated integrally, at an air ratio of 1.2, as it is customary for industrial firing systems operating atmospherically. In contrast to the differential tests presented above, reactions of alkali capture in the ash and slag





**Fig. 7.39** Evaporation of sodium and potassium for different coal types and concentrations in the gas phase as a function of the particle temperature (Aho et al. 1995)

could be observed. This means that a clear decrease of alkali concentrations occurs as the residence time in the reactor increases. Figure 7.40 shows the measured concentrations for different hard and brown coals. The maximum rate of release in the tests for the Ensdorf hard coal, at 1,400°C and a short residence time, was about 10% for sodium and about 5% for potassium. With longer residence times, a high capture in the ash was observed, entailing a reduction of the gaseous alkali concentrations of more than one order of magnitude. The capturing effect in the ash was significantly weaker with brown coal due to the lack of aluminium silicates. By



**Fig. 7.40** Gas-phase sodium and potassium concentrations for combustion of different coal types (Reichelt 2001)

injecting additives such as alumina and silica in the combustion of the Ensdorf coal, reductions up to one order of magnitude were achieved (Schürmann et al. 2001).

#### 7.4.3.3 Secondary Alkali Removal

For pressurised pulverised coal firing, the removal of alkali compounds is a prerequisite to preventing damage of the gas turbines. The low temperatures of pressurised fluidised bed combustion involve lower concentrations of volatile alkali compounds, and what is more, the turbine is less sensitive to alkali loading because of the low gas turbine entry temperature. The PFBC plants currently in service operate without alkali removal. For hybrid pressurised fluidised bed processes, however, the higher gas turbine entry temperatures entail similar requirements to pressurised pulverised coal firing.

The abatement of alkali concentrations hazardous to the turbine can be achieved by a combination of methods, proposed as follows:

- Leaching of the coal to reduce the contents of alkali chlorides.
- In situ absorption in the furnace by injection of additives and selection of process parameters to raise the rate of capture.
- Absorption in a filter downstream of the combustion. A packed-bed filter, for instance, besides having the function of particle separation can in addition be used for alkali removal (see Sect. 7.3.2).

The most preferable method of alkali removal is to pass the combustion gases through a fixed bed of non-volatile inorganic solid sorbents. Potential sorbents should show a high temperature stability, fast sorption kinetics and a high loading capacity. Kaolin, bauxite and emathlite have been identified as suitable sorbents for gas-phase alkalis. The capturing mechanism can be based upon the principle of physical absorption and/or upon a chemical reaction.

In the temperature range of fluidised bed firing systems, the most effective removal process is physical absorption of condensed alkali components followed by chemical bonding. In contrast, given the high temperatures, the chemical reactions are most effective in pressurised pulverised coal firing. A chemical reaction consumes the getters, while exclusively physical absorption allows their regeneration. As physical absorption is a reversible reaction and alkalis could be released by pressure and/or temperature changes, chemical reactions are the preferred option.

Studies into the suitability of potential getter materials have for the most part used synthetic gases at temperatures in the range of fluidised beds (Punjak et al. 1989; Uberoi 1990; Turn et al. 1998). Only recently have results for higher temperatures become available (Oleschko and Muller 2007; Willenborg et al. 2006; Escobar and Muller 2007; Escobar et al. 2008).

In another investigation, using a portion of the flue gas flow from a PFBC plant, different sorbents in a granular bed filter were tested for alkali removal. The purpose of these tests was to develop an alkali measurement technology employing absorption of the alkalis in a bulk bed, with subsequent analysis of the absorbed alkalis. Measurements at bulk bed temperatures of 800–850°C and a pressure of 9

bar showed an 88% removal of the gaseous sodium using activated bauxite and an 86% removal with diatomaceous earth (Thambimuthu 1993; Lee and Carls 1990; Lee and Swift 1991).

At a laboratory-scale PFB furnace with particulate and alkali removal, data was obtained in practical tests with a packed-bed filter (Zakkay et al. 1989). During the tests, the plant was operated at a temperature of between 809 and 871°C and a pressure of between 7 and 9.3 bar. The particle- and alkali-laden flue gas from the PFB furnace flowed into a granular bed filter after an initial cleaning step in a cyclone. The filter medium was of alumina granulates. The measurements showed an average dust removal degree of 97.8–99% and an alkali removal degree of 90–96%. The alkali emissions at the outlet of the granular bed filter ranged between 0.0019 and 0.041 mg/kg for sodium and between 0.001 and 0.031 mg/kg for potassium, which meant most of the measurements were below the limit of 0.024 mg/kg given by gas turbine manufacturers.

Documentation of industrial-scale experience with alkali capture by such a filter arrangement is not available.

Studies into alkali capture at temperatures above 1,300°C are being carried out at a 1 MW<sub>th</sub> experimental plant within the framework of the joint pressurised pulverised coal combustion project. The subject of the investigations is the capture of gaseous alkalis by the molten coal slag either in the furnace or in the downstream packed-bed filter for molten ash removal. Given that the getter materials silicon and aluminium oxide are the main components of coal ash, it seems reasonable to make use of this getter potential. The main influencing parameters in alkali removal by slag are the temperature and, to a great extent, the residence time (Hannes 1996).

In tests at the 1 MW PPCC furnace, considerable reductions of the alkali concentrations were achieved, supporting the theory of the molten ash removal process. Starting out from an initial 18 mg/m<sup>3</sup>, the alkali concentrations were reduced to about 8 mg/m<sup>3</sup> by using inertia separators and reduced further to less than 6 mg/m<sup>3</sup> by fine particulates removal. The downstream alkali removal by non-optimised aluminosilicate sorbents affected another clear reduction to less than 2 mg/m<sup>3</sup>. Because vaporous alkalis are present in an ionised state at high temperatures, the alkali capture by means of slag or getters can be improved by the implementation of electrical fields. This has been proven in laboratory-scale investigations at high temperatures of 1,300°C (Hübner et al. 2003). By also using electrical fields, the alkali concentrations in the 1 MW PPCC furnace could be reduced to 0.2 mg/Nm<sup>3</sup> (Förster et al. 2005; Müller et al. 2008).

Accompanying fundamental investigations at lab scale were carried out to determine the alkali retention potential of the coal ash present in the combustion chamber and the liquid slag separator. For this reason, alkali partial pressures over coal ash slags with and without additives were determined by Knudsen effusion mass spectrometry (KEMS). Although alkali vapour reductions of one order of magnitude could be achieved by the addition of SiO<sub>2</sub> or TiO, the vapour pressures are still about 2 orders of magnitude higher than those required by gas turbine manufacturers. Therefore, a separate alkali removal unit is necessary to achieve alkali concentrations in the hot flue gas compatible with these specifications. In

further investigations, model sorbents were exposed to sodium-containing gases. Aluminium silicates having an  $\text{Al}_2\text{O}_3/\text{SiO}_2$  ratio of about 1:8 showed the highest sorption capabilities (Willenborg et al. 2006). In laboratory-scale flow channel experiments at  $1,400^\circ\text{C}$ , kaolin and silica-enriched bauxite have shown the best potential for sufficiently removing the alkalis. The alkalis are bound in a glass melt state formed during alkali sorption. The total NaCl concentration can be reduced to less than 30 ppbv using kaoline or silica-enriched bauxite. The sorption reaction of potassium is favoured over that of sodium. However, with sufficient water, both reactions take place to equal extents (Escobar and Muller 2007).

Thermodynamic calculations based on the experimental results were used for a first estimation of the risk of hot corrosion to the turbine blading of a PPCC (Escobar et al. 2008; Müller 2008). By expanding and cooling the flue gas in the gas turbine, alkali components can condense and cause corrosion. For corrosion to occur, two conditions are necessary:

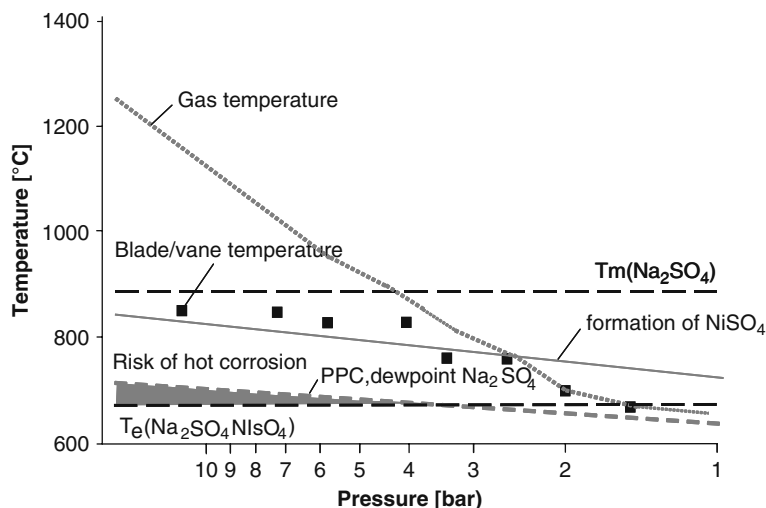
- The components have to condense, which means that the blade temperatures must be below the dew point or condensation temperature.
- The dew point has to be higher than the melting temperature, because corrosion is caused by liquids.

Hot gas corrosion (type I) is caused by the formation of liquid  $\text{Na}_2\text{SO}_4$  above its melting point ( $884^\circ\text{C}$ ). Another type of hot gas corrosion (type II) is caused by the formation of a eutectic melt of  $\text{NiSO}_4$  and  $\text{Na}_2\text{SO}_4$  above  $671^\circ\text{C}$ .  $\text{NiSO}_4$  is formed by the reaction of the oxide scale of Ni based alloys with  $\text{SO}_3$  according to the  $\text{SO}_3$  partial pressure in the hot flue gas. Therefore, the coexistence of  $\text{Na}_2\text{SO}_4$  and  $\text{NiSO}_4$  or the formation of liquid  $\text{Na}_2\text{SO}_4$  is taken as the criterion for the risk of hot gas corrosion.

The thermodynamic stability of the sulphates and other species in the gas turbine was calculated using a three-staged reactor model consisting of a combustion chamber, hot gas cleaning and a gas turbine. The thermodynamic equilibrium was calculated for all stages. In the hot gas cleaning stage, the alkali content of the gas was set to 2 ppbv at 16 bar according to the experimentally obtained values of about 24 ppbv at 1 bar. The ultimate thermodynamic stability of the sulphates depended on the equilibrium calculation of the gas turbine stage.

The results of the thermodynamic calculations are shown in Fig. 7.41.  $\text{Na}_2\text{SO}_4$  is the first alkali species that condenses – coal contains high amounts of sulphur and  $\text{Na}_2\text{SO}_4$  is the least volatile sodium species in the flue gas. The condensation temperatures of  $\text{Na}_2\text{SO}_4$  range from  $650^\circ\text{C}$  at 1 bar to  $720^\circ\text{C}$  at 16 bar. These temperatures are below the melting temperature. Furthermore, there should be no condensation of  $\text{Na}_2\text{SO}_4$  on the gas turbine blades at all, because the calculated dew point is lower than the temperature of each blade. Therefore, no hot gas corrosion (type I) should take place. The dew point temperature of  $\text{K}_3\text{Na}(\text{SO}_4)_2$ , the thermodynamically most stable potassium sulphate, is 20 K lower again, so the corrosion risk is even lower.

The shadowed area marks the region in which both  $\text{Na}_2\text{SO}_4$  and  $\text{NiSO}_4$  are stable above the eutectic temperature of  $671^\circ\text{C}$ , where hot gas corrosion type II may occur.



**Fig. 7.41** Results of thermodynamic calculations for the estimation of hot corrosion risks (from Escobar et al. 2008, © 2008, with permission of Elsevier)

However, operation of the blades does not occur at these critical conditions (Escobar et al. 2008; Müller 2008).

## 7.4.4 State of Development

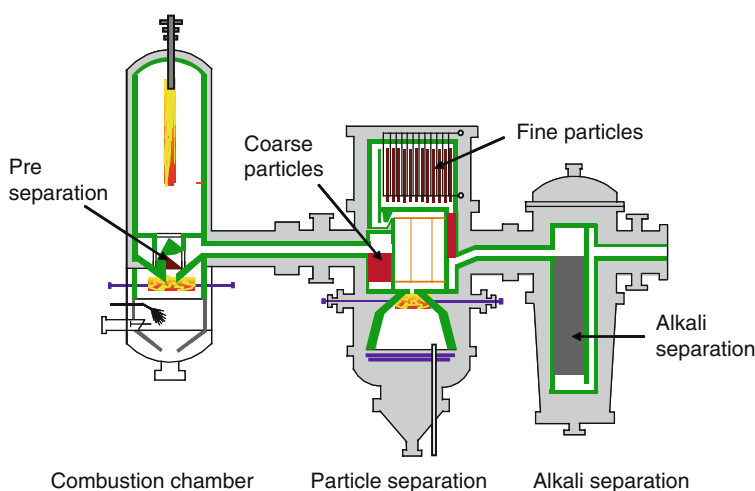
### 7.4.4.1 Germany, Pressurised Pulverised Coal Combustion Project

Following preliminary investigations into cleaning hot flue gases from a small-scale pressurised pulverised coal combustion (PPCC) furnace at the *Institut für Energie- und Umweltverfahrenstechnik der Universität Duisburg-Essen (IUVT)*, “Institute for Energy and Environmental Protection Technologies, Duisburg-Essen University”, the process of PPCC was investigated and developed by several German industry partners from 1989 to 2005. The objective of the joint project was the production of hot gases from a PPCC furnace suitable for gas turbines. The various planned and accomplished steps of development are compiled in Table 7.11. The project was discontinued after 2005, and a pilot-scale plant was not constructed.

Within the framework of the joint project, a test plant using PPCC with a thermal capacity of 1 MW was erected and operated in Dorsten, Germany. The test plant comprised of the furnace, a molten slag and alkali removal unit and a testing section for turbine blade materials. The schematic of the test plant is shown in Fig. 7.42, while Fig. 7.43 shows the combustion chamber with the fly ash and alkali removal stages. The furnace was designed for a temperature of 1,700°C and a pressure of up to 20 bar. The construction was an uncooled multiple-leaf refractory-lined upright chamber with a roof burner and bottom outletting of slag. Slag flowing down the

**Table 7.11** PPCC Development Programme (Förster et al. 2005)

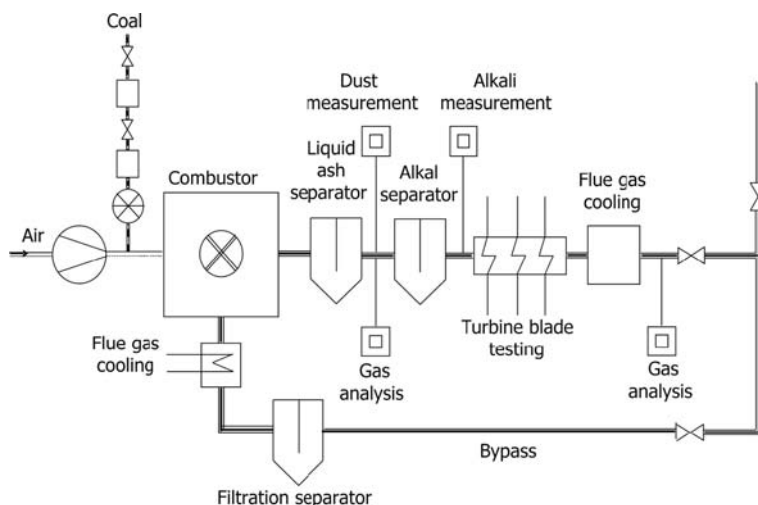
Phase	Planned development	Period
1	Planning, construction and putting into service of a 1 MW <sub>th</sub> test-scale plant in Dorsten, North Rhine-Westphalia	1989–1992
2	Selection and development of suitable materials resistant to molten ash for the construction of the furnace and the molten ash separator	1993–1995
3	Development of the molten ash and alkali separator	Since 1996
3a	Investigation of the various inertia separators	1996–2002
3b	Development of the fine particulates and alkali separation technologies	2003–2005
4	Planning, construction and operation of a pilot-scale plant (10 MW <sub>th</sub> capacity); operation of the demonstration plant	–

**Fig. 7.42** Schematic drawing of the 1 MW PPCC facility (Förster et al. 2005)

chamber walls and over the floor granulated in a water bath. The slag-tap furnace was followed by a flue gas cleaning unit with molten slag and alkali removal. Integrated into the flue gas duct, a testing segment served to investigate the behaviour of different turbine blade materials in the flue gas (Hannes 1986; Preußner and Spindler 1988; Förster et al. 2001; Hannes 2002; Förster et al. 2005).

The results of the investigations into particle and alkali removal have been presented in Sects. 7.4.2 and 7.4.3.3. As a result of many years of development, it became possible, after separating coarse and removing fine particles, to ascertain particle concentrations below 1 mg/m<sup>3</sup> for particles smaller than 3 µm. By doing so, the strict requirements of modern gas turbines could be complied with (Förster et al. 2005).

As far as alkali removal is concerned, it is assumed that, by the primary capture of alkalis in the molten ash inside the combustion chamber and by a secondary cleaning



**Fig. 7.43** 1 MW PPC combustion chamber and hot gas cleaning (Förster et al. 2005)

step, it will be possible to comply with the required limit values of  $0.01 \text{ mg/m}^3$  (Förster et al. 2005).

Considerable work within the PPCC project was also directed towards testing and developing materials suitable for the high-temperature zones of the combustion chamber, the molten ash separator and the alkali removal unit. Such materials should have a high temperature stability, resistance and density to withstand the molten slag. Suitable materials are isostatically pressed or fusion-cast ceramics. They are temperature and corrosion resistant and have a high density and low porosity, which prevent the slag from penetrating the refractory lining (Weber et al. 1993). A drawback, however, is the high chromium content, which evaporates at the temperatures used, forming, in particular, compounds with alkalis, which is likely to mean deposits in the gas turbine. Potential solutions to the problem are coatings to reduce the evaporation of the chromium or the use of hafnium oxide based ceramics, which are free from chromium (Förster et al. 2001; Müller 2008).

#### 7.4.4.2 Efficiency Potential and Design of PPCC Furnaces

The joint PPCC project was the framework for the planning of industrial-scale plants with capacities of 150 and 300  $\text{MW}_{\text{el}}$  (as of 1992). The configuration of the 150  $\text{MW}_{\text{el}}$  PPCC furnace was a two-line concept, each line with a combustion chamber and a gas turbine. The type of gas turbine taken as a basis was an existing gas turbine, V64.3 by Siemens, with a capacity of 53  $\text{MW}_{\text{el}}$  at an ISO gas turbine entry temperature of  $1,150^\circ\text{C}$ . Each combustion chamber was designed for a thermal output of 159 MW. The combustion chamber had an inner diameter of 3 m and a length of 6 m. The waste heat recovery process was shared by the two lines and was executed as a dual-pressure (steam) cycle, with moderate steam conditions of 70 bar



and 535°C. The efficiency of the concept plant was 48%. An electrical output of 300 MW<sub>el</sub> could be produced by doubling the number of modules. Depending on the cogeneration process quality, it is possible to achieve efficiencies of up to 51% (Leufert 1993).

For the industrial-scale design of a combustion chamber, two variants are distinguished, as in the case of pressurised fluidised bed furnaces. The combustion chamber can be designed either with water – steam cooling, like in conventional atmospheric furnaces, or without cooling for adiabatic operation. From the thermodynamic point of view, the adiabatic combustion chamber has advantages because the total fuel heat can be fed to the gas turbine by this technology. With an air ratio of about 1.7, temperatures are limited to around 1,600°C in the combustion chamber, and gas turbine entry temperatures are achieved by adding additional air. Using adiabatic hot gas production, the resulting gas-to-steam turbine output ratio is 65:35, roughly similar to a natural gas fired cogeneration process. The advantage of water – steam cooling is the thermal design flexibility and gas tightness of the wall. Using this method, cooling of the flue gases is achieved by heat dissipation to the membrane walls, not by setting a high excess air level, as in adiabatic firing. The ratio of the gas-to-steam turbine output then diminishes to 55:45, while the efficiency decreases from 48 to 44%. In practice, though, it will be advisable to opt for a middle course where the wall thicknesses of the refractory linings are reduced by the use of a certain amount of cooling. This way, the plant has a better thermal flexibility; it stores less heat and has shorter heat-up and cooling-down times (Preußner and Spindler 1988; Reichert et al. 1988).

Recent calculations of PPCC systems give overall efficiencies between 53.3 and 55%, using higher gas turbine entry temperatures of 1,200–1,400°C and pressure conditions optimised to these temperatures. Extensive studies using various parameters were carried out with a view to analysing pressure and heat losses from the hot gas cleaning process and the combustion chamber cooling. The results are compiled in Table 7.12 (Schuknecht 2003).

The efficiency of the PPCC process is about 6% lower than that of the natural gas fuelled combined cycle process at the same gas turbine inlet and waste heat

**Table 7.12** PPCC cycle calculations (Schuknecht 2003)

Turbine inlet temperature (ISO)	°C	1,200	1,300	1,400
Optimal gas turbine pressure ratio	–	16	17	30
GT exit temperature	°C	583	637	622
GT flue gas mass flow	kg/s	628	632	635
Live steam temperature	°C	565	619	604
Live steam pressure	MPa	18	18	18
Boiler exit temperature	°C	140	140	140
Fuel input (LHV)	MW	632.1	753	855.2
Power – gas turbine	MW	220.7	259	299
Power – steam turbine	MW	131.3	172	194.3
Auxiliary power	MW	13.7	16.1	17.1
Electrical net power	MW	338.4	414.9	476.2
Efficiency (LHV)	%	53.5	55.1	55.7

steam generator conditions. The efficiency loss in comparison to a natural gas fired combined cycle is due to several reasons:

- In the combined process with PPCC, the auxiliary power demand, of about 4% of the gross electric power generation, is about double that of the natural gas fuelled process, due to the power needed for coal preparation, flue gas desulphurisation and  $\text{DeNO}_x$ . This causes an efficiency decrease of 2% in comparison to a gas-fired combined cycle.
- If the heat loss of the combustion chamber and the hot gas cleaning unit is 0.5%, accordingly the efficiency decrease is then about 0.5%.
- The pressure loss in the combustion chamber and the gas cleaning unit decreases the efficiency by approximately an additional 0.5%.
- Water/steam cooling of the combustion chamber further decreases the efficiency. The larger the heat flow dissipated to the steam turbine using combustion chamber cooling, the higher the efficiency loss.
- Another reason for the efficiency difference is the higher flue gas exit temperature needed when using coal as the fuel (Preußner and Spindler 1988; Schuknecht 2003).

#### 7.4.4.3 USA

In the USA, the Westinghouse, Solar Turbines and Allison Gas Turbines companies developed processes with coal-fuelled gas turbines supported by the Department of Energy (DoE). While Solar and Allison wanted to provide plants with a capacity of  $20 \text{ MW}_{\text{el}}$  for industry, Westinghouse focussed on the power plant sector, with capacities of  $200 \text{ MW}_{\text{el}}$ .

The principal configurations of these variants were shown previously, in Fig. 7.32. The gas turbine entry temperature of all of the process variants was only around  $1,000^\circ\text{C}$ , which limits the efficiency. The reasons for developing these processes was cost reduction (in comparison to conventional power plants) and the ability to construct small-capacity units using coal as fuel (Parsons and Byam 1989).

The processes were investigated at pilot scale up until 1993; publications on newer investigations do not seem to be in the public domain. It seems reasonable to assume that, due to operational problems (especially with hot gas cleaning), PPCC development has been discontinued in the USA. Publications in the USA to do with new coal-firing concepts do not involve PPCC, which supports this assumption.

#### 7.4.4.4 Westinghouse

The Westinghouse Company carried out investigations at a furnace with a capacity of  $3.5 \text{ MW}_{\text{th}}$ . The schematic diagram of the furnace is shown in Fig. 7.44. The process concept is based on two-stage combustion, molten slag removal and low gas turbine entry temperatures of  $1,000^\circ\text{C}$ , controlled by a high excess air level in the second combustion stage.

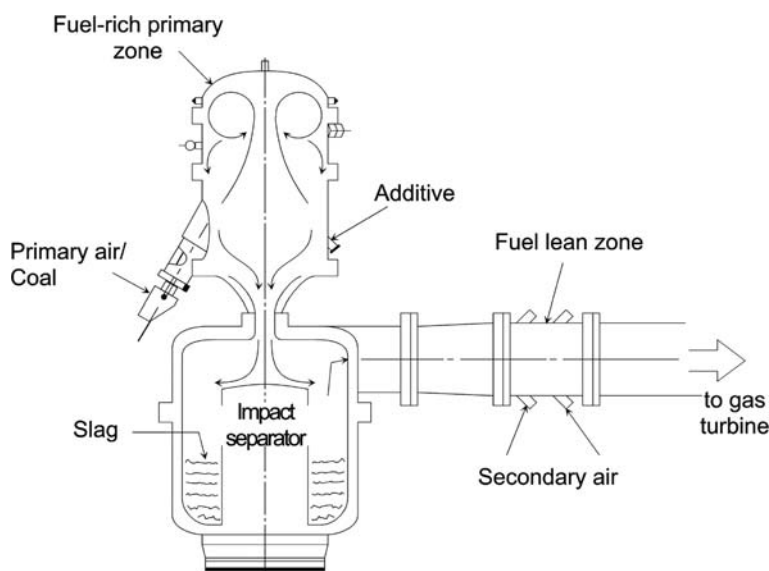


Fig. 7.44 Westinghouse's PPCC facility (Pillsbury et al. 1989)

The slag-tap furnace was operated under a pressure of 6 bar, at a temperature of  $1,600^{\circ}\text{C}$  and an air ratio of 0.7. This way 98–99% of the carbon was converted into the gas phase, i.e. into  $\text{CO}$  and  $\text{CO}_2$ . Four burners directed upwards at an angle combined to produce a single flame shape, in the process forming a swirl, deflected downwards from the top of the furnace. This flow pattern ensured both stabilisation of the flame and a high ash retention.

During firing, a slag layer would form on the chamber walls, flowing by gravity towards the bottom of the chamber, where lime or limestone was added for capture of sulphur oxides. An impingement separator followed after the bottom of the chamber. Here, the flue gases of the primary combustion chamber were sped up by an outlet nozzle so that ash particles, while accelerating, impinged and stuck to the separator. Its geometrical design acted in such a way that particles greater than  $5\text{ }\mu\text{m}$  were separated. Molten slag flowed down the walls of the separator. In test operation, a total ash removal of more than 90% could be achieved.

By adding secondary air, the burnout, especially of the gas phase, was attained in the secondary combustion chamber. The total air ratio was set at 2.5 to achieve a cooling to the desired gas turbine entry temperatures.

Using two-stage combustion, it was possible to achieve  $\text{NO}_x$  emissions below  $250\text{ mg/Nm}^3$  (Bannister et al. 1990). The stated sulphur retentions ranged from 30 to 40% at a  $\text{Ca/S}$  ratio between 2 and 4. Analyses of the slag showed that about 80% of the alkalis were captured in the slag (Pillsbury et al. 1989). After the secondary combustion chamber, about 1–2% of the alkalis from the coal were still found in the gas phase. Later, an additional cyclone was installed in the test plant between the impingement separator and the secondary combustion chamber. This way, the total ash removal was increased to 99% (Bannister et al. 1992).

Studies discussed drafts for a 200 MW<sub>el</sub> plant. Featuring a low gas turbine entry temperature of 1,010°C and a corresponding low turbine exit temperature of 370°C, a steam turbine output of 50 MW<sub>el</sub> and a 160 MW<sub>el</sub> output from the two gas turbines, the stated total efficiency was 37.5%. The concept was meant to give 20% cheaper electric power production compared to a conventional coal-fuelled power plant with flue gas desulphurisation. A precondition to that estimate, though, was that additional hot gas cleaning was not necessary (Pillsbury et al. 1989). Difficulties with the flue gas cleaning in the slag-tap furnace and in the secondary combustion zone and, more obviously, the efficiency of 37.5% not being commensurate with the set target led to a change in the concept. In a later publication, a modified concept was presented (Bannister et al. 1992) which differed from the PPCC concept, including gasification process elements as well as externally fired gas turbine process features.

#### 7.4.4.5 Solar Turbines

After several preliminary investigations, Solar Turbines developed a pulverised coal furnace for a 3.8 MW<sub>el</sub> gas turbine (Cowell and LeCren 1992; Cowell et al. 1992b). The schematic drawing of the pilot-scale plant is shown in Fig. 7.45. It consisted of a first combustion stage, an uncooled, adiabatic slag-tap furnace, a particle separator and a second combustion stage. The fuel was a coal – water suspension. The first combustion stage was operated at air deficiency (an air ratio of 0.7) and high temperatures of 1,600°C. The combustion pressure was 9.6 bar. The furnace was followed by a molten slag separation unit. Additional air injection completed the combustion in a second stage, with the flue gases being cooled to 1,040°C in the process. The total air ratio ranged around 2.5. Before the cleaned hot gas entered the gas turbine,

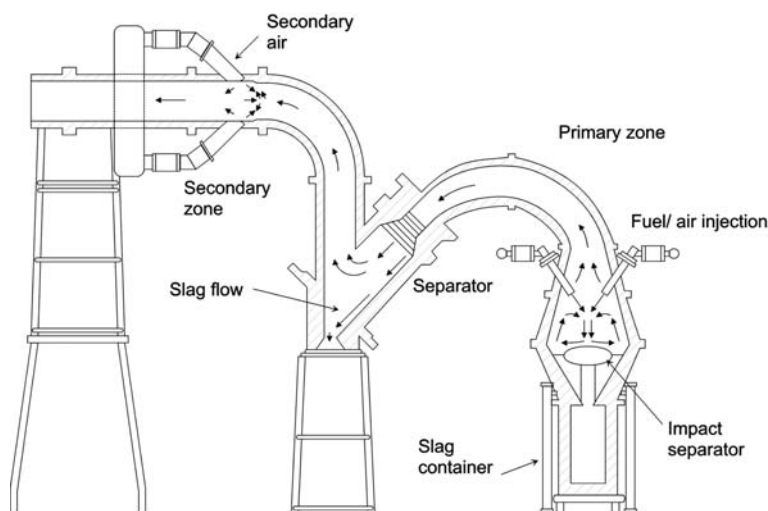


Fig. 7.45 Solar Turbines' PPCC facility (Cowell et al. 1992b)

the plant had another dust removal unit, using ceramic filters, at the secondary combustion temperature (Cowell et al. 1992a).

The separation unit installed between the combustion chamber and secondary air injection consisted of seven staggered rows of ceramic rods with a diameter of 3.2 cm, arranged with a clearance of 1.9 cm. The flue gas duct cross-section was widened to reduce the flow velocity, thus preventing the ash removed by the rods from being re-entrained (Cowell et al. 1992b).

During the investigations at the test plant, an ash removal of 61% in the combustion chamber and of 98% after the ceramic rods between the primary and secondary zones could be achieved. The particle content was thus about 40 mg/kg flue gas. The staged combustion made it possible to achieve low  $\text{NO}_x$  emissions of 150 mg/Nm<sup>3</sup> at 6%  $\text{O}_2$  (calculated as  $\text{NO}_2$ ) along with equally low CO emissions of 30 mg/Nm<sup>3</sup> at 6%  $\text{O}_2$ . There are no results available from the operation of the pilot-scale plant, including for the ceramic filters and the gas turbine.

#### 7.4.4.6 Allison

The concept of Allison differed from the concepts of Westinghouse and Solar by not including slag removal. Only dry fly ash was separated, at temperatures around 1,000°C. The coal – water suspension was burned in a dry bottom furnace at temperatures of 1,200°C, at air deficiency. The flue gases were cooled to below 1,000°C by a water quench; the fly ash was removed in a cyclone. After a second combustion stage, with injection of the remaining combustion air, and another cyclone stage for fly ash removal, the hot gases were fed to the gas turbine (Parsons and Byam 1989).

Allison carried out tests with a 3.5 MW<sub>el</sub> gas turbine firing coal at part load so that an entry temperature of 816°C was achieved. After each 4 h test, the gas turbine was unmounted and the blading inspected. A mass balance revealed that about 0.2% of the fuel ash had been deposited in the turbine. An analysis of the deposits indicated an enrichment of alkalis and sulphur. The deposits could be easily removed from the blades by washing, but with longer operation, it would be expected that adhesive, stable deposits of ash would form. In addition, corrosion would occur. As a result, alkali separation (from the flue gas) would have to be introduced into the process for any practical applications. The authors wanted to achieve this by a further lowering of the quench stage temperature.

#### 7.4.5 Summary and Conclusions

PPCC offers the potential for a high efficiency in comparison to the competing coal-based combined cycle processes. To exploit this potential, high-temperature particle and alkali removal at temperatures of 1,400–1,600°C is a prerequisite. Although substantial progress in both fields has been achieved, the PPCC development in Germany and, it seems, in the USA, has been discontinued.

## 7.5 Externally Fired Gas Turbine Processes

Combined cycles with integrated coal gasification or pressurised pulverised coal firing use a gas cleaning stage to produce a clean fuel (i.e. a hot gas) for the gas turbine. The externally fired gas turbine process, in contrast, uses a heat exchanger to heat a gas already clean enough to meet the gas turbine related requirements. This indirectly fuelled or externally fired combined cycle (EFCC) presents an alternative to the combined cycle processes that integrate coal gasification or operate using pressurised firing (Baum 2001; Spliethoff 2000; Spliethoff and Baum 2002a, b; Benson 2000).

### 7.5.1 Structure, Configurations, Efficiency

In an indirectly fired combined cycle, the heat from the fuel is released by combustion in a firing system. A heat exchanger follows the furnace, the hot flue gases giving heat to a clean, pressurised turbine working fluid. Hot gas cleaning is not needed for the indirectly fired process, as the flue gases and turbine fluid are segregated by the heat-exchanging walls. Therefore, any necessary flue gas cleaning to meet emission limits can be performed at low temperatures, as in conventional coal-fired power plants.

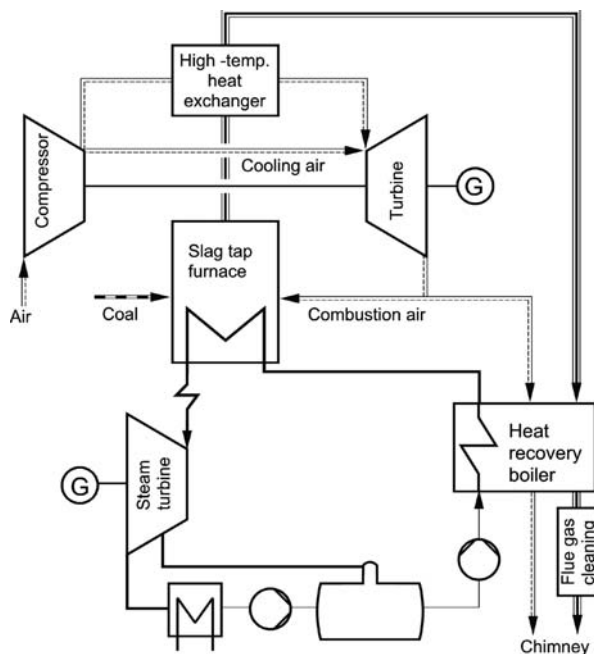
There are various configurations possible for indirectly fired gas turbine combined cycles. Open gas turbine processes use air or cleaned flue gas as the turbine working fluid, while closed gas turbine processes work with gases such as helium or carbon dioxide. These gases have better thermodynamic properties than air or hot flue gas but can only be used in closed gas turbine processes.

Figure 7.46 shows a schematic diagram of an open process using air as the working fluid for the gas turbine. After compression, the air is heated to the gas turbine inlet temperature in the high-temperature heat exchanger. Part of the gas turbine exhaust air is fed to the firing as combustion air. The heat of the remaining exhaust air is used in a heat recovery process together with the remaining hot flue gas heat which was not transferred in the high-temperature heat exchanger.

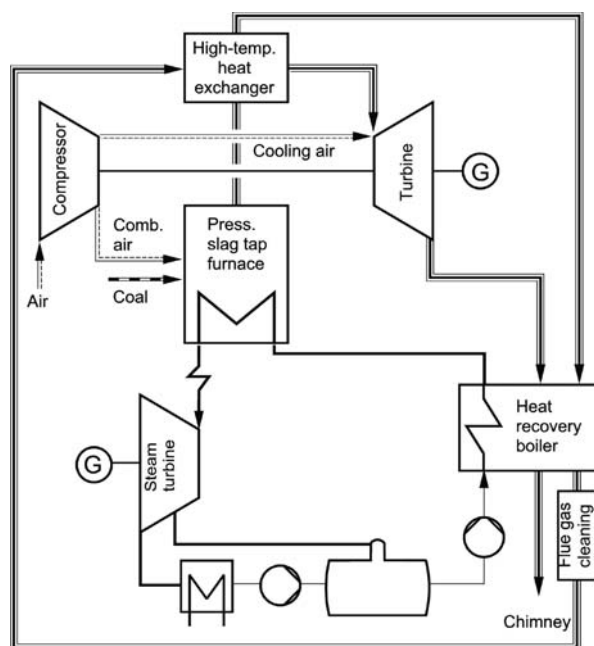
The configuration of an open EFCC process using flue gas as the working fluid for the gas turbine shown in Fig. 7.47 employs pressurised slag-tap firing. The hot pressurised flue gas transfers heat in a high-temperature heat exchanger and in a heat recovery process. It is cleaned at a low temperature before being reheated in the high-temperature heat exchanger, then enters the gas turbine. In contrast to the open variant using air, this process has the advantage of not subjecting the heat exchanger to compressive stresses except through its own pressure drop and that of the gas cleaning process. Equipment costs, however, are significantly higher than using air as the working fluid for the gas turbine.

Figure 7.48 shows a schematic diagram of a closed EFCC process. The working fluid of the gas turbine is conducted in a closed circuit, while the firing and the flue gas path remain unmodified. The basic advantage is the potential to use gases with better thermodynamic properties for the heat transfer. The high compressor inlet temperature, however, has a negative effect on the overall efficiency.

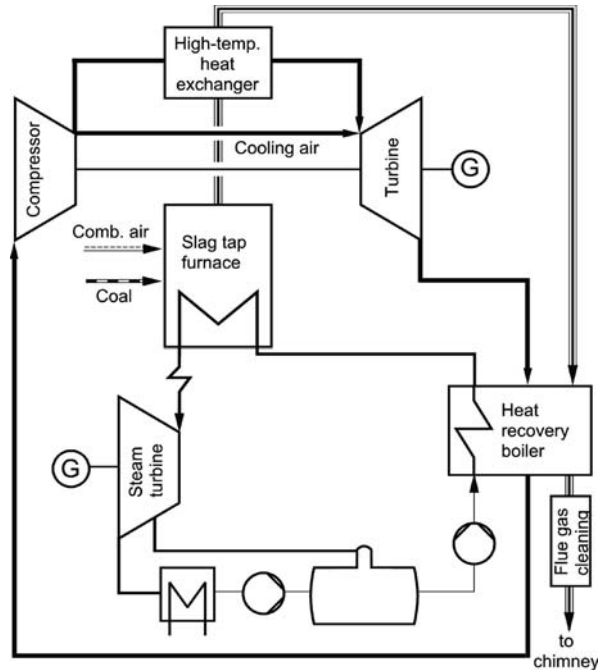
**Fig. 7.46** An open EFFCC process using air (atmospheric slag-tap furnace) (Spliethoff and Baum 2002)



**Fig. 7.47** An open EFCC process using flue gas (pressurised slag-tap furnace) (Spliethoff and Baum 2002)



**Fig. 7.48** A closed EFCC process (atmospheric slag-tap furnace) (Spliethoff and Baum 2002)



The EFCC process with auxiliary natural gas firing shown in Fig. 7.49 is analogous to a natural gas fired combined cycle with combustion air preheating by coal flue gas in a high-temperature heat exchanger. In this process, this exchanger is not subjected to the high thermal stress as in the variants described so far. The coal firing need not necessarily use a slag-tap furnace. The type of furnace depends on the temperature the heat exchanger operates at. The natural gas demand decreases with an increasing temperature.

The following material will refer to open gas turbine processes using air as the working fluid because this configuration is rather uncomplicated while having potential for a high efficiency. Most of the data presented, however, is applicable to the other EFCC configurations as well.

The efficiency of the EFCC process essentially depends on the following parameters:

- The heat exchanger outlet and the gas turbine inlet temperatures
- The cooling of the furnace
- The cycle efficiency of the steam generation process
- The flue gas losses

The rest of this section (Sect. 7.5.1) shall illustrate, using calculations over the cycle, the effect on the efficiency of the major parameters. The baseline case of these calculations is shown in Fig. 7.50, a power plant with an electrical capacity of 350 MW (Baum 2001).



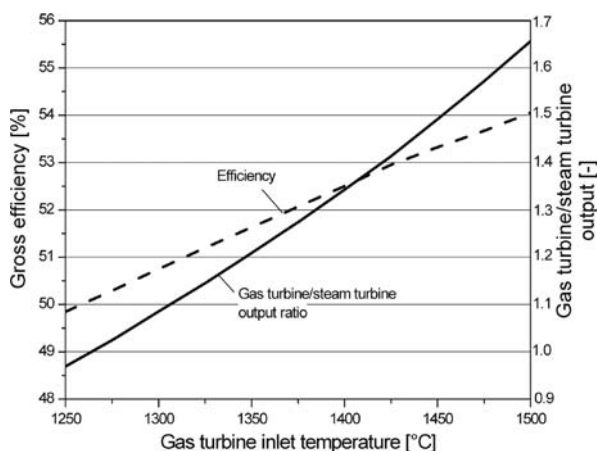


The input of heat from the fuel (coal) to the slag-tap furnace is 700 MW in the model case. The furnace uses steam – water cooling, and about 7% of the total fuel heat input is directly transferred to the steam – water cycle. Further cooling to the flue gas outlet temperature of 1,600°C is achieved by operating at an air ratio of 2.1. In the high-temperature heat exchanger which follows, the flue gas transfers its heat to the compressed air, heating it to 1,375°C.

In order to cool the gas turbine, part of the compressor air is conducted directly to it. The result is an ISO gas turbine inlet temperature of 1,184°C for this baseline case. That part of the gas turbine exhaust which is not used as secondary air is either used for the drying-milling process of the coal, and for its transport, or is mixed with the flue gas from the combustion of the coal and used for waste heat steam generation.

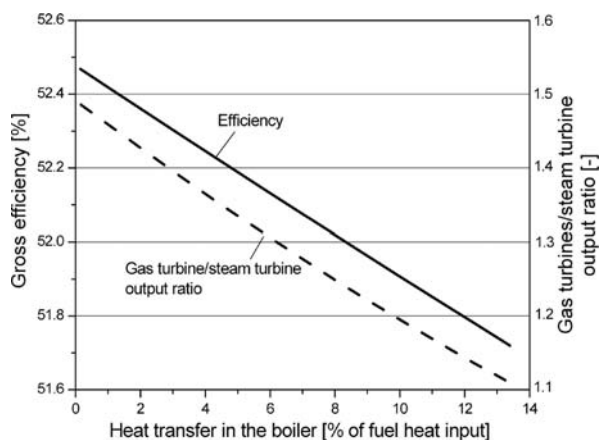
Taking into account the auxiliary power demand for coal preparation, pumps and the induced-draught, and other losses, totalling about 6% of the gross electrical output, the gross efficiency of 52% is reduced to a net efficiency of 49% for the baseline case. It should be mentioned that, given the conservative assumptions about the machine efficiency, the terminal temperature difference in the waste heat boiler and so forth, the process discussed here is not efficiency optimised. The efficiency potential of the EFCC process is comparable to the IGCC process. With higher ISO turbine inlet temperatures of 1,250°C and an improved steam cycle it should be possible to achieve a net efficiency of about 52%, comparable to the IGCC 98 case (see Sect. 7.6) (Edelmann and Stuhlmüller 1997).

The influence of the heat exchanger outlet temperature (which is to say the real gas turbine inlet temperature) on the efficiency of an EFCC process is shown in Fig. 7.51. The heat exchanger outlet temperature and the thermodynamic efficiency increase with higher mean heat input temperatures. The gas turbine share of the total process output increases as well. At a constant mass flow, higher gas turbine inlet temperatures make larger heat transfer surfaces necessary. As the high process temperatures make it impossible to use metal, it is only possible to use ceramic



**Fig. 7.51** Efficiency and the gas turbine/steam turbine output ratio as a function of the real gas turbine inlet temperature (Spliethoff and Baum 2002; Baum 2001)

**Fig. 7.52** Influence of furnace cooling on the efficiency and the gas turbine/steam turbine output ratio (Baum 2001)



materials for these surfaces. The ceramic heat exchanger is the only component in the indirectly fired combined cycle which is not yet state of the art. It is discussed in detail in Sect. 7.5.2.

The cooling technique for the furnace has a substantial influence on the EFCC process, in a similar way as in pressurised pulverised coal firing (see Fig. 7.52). The efficiency is highest if the heat of the furnace and heat exchanger is transferred to the working fluid of the gas turbine and the steam-generating process is heated only by the waste heat from the gas turbine.

This can be achieved by an adiabatic furnace without cooling or by an air-cooled furnace. Water/steam cooling, as a third option, is simpler to construct. It was therefore chosen as the basis for calculations.

The cycle efficiency of the steam process is another important parameter influencing the total efficiency of the EFCC process. Since the steam process contributes 40 – 50% of the total output, depending on the type of furnace cooling and the gas turbine inlet temperature, an improvement of the steam process by 1% means an increase of the total efficiency of 0.4 – 0.5%.

## 7.5.2 High-Temperature Heat Exchanger

The concept of an indirectly fired gas turbine is an alternative to combined cycle processes with integrated coal gasification or pressurised firing which have enjoyed the majority of research efforts. A critical aspect of the indirect firing concept if it is to be put into practice is the high-temperature heat exchanger where the heat of the flue gas is transferred to the working fluid of the gas turbine.

### 7.5.2.1 Requirements

The requirements in process-engineering terms for such a heat exchanger are the following:

- **Process temperatures of up to a maximum of 1,600°C**

Modern gas turbines work at ISO gas turbine inlet temperatures of up to 1,300°C, which correspond to real gas turbine inlet temperatures of up to 1,500°C. To achieve these temperatures on the side of the cleaned gas of the high-temperature heat exchanger, it is necessary to be able to set flue gas temperatures of up to 1,600°C on the side of the untreated gas.

- **Compressive stress of the heat exchanger**

In the case of the open EFCC process using air, the heat-transferring walls are subjected to stress by the pressure difference between the turbine working fluid and the flue gas. This difference is the result of the compression ratio of the gas turbine, which is around 16–18 for gas turbines designed for natural gas with gas turbine inlet temperatures of 1,200–1,300°C. It must be observed in this respect that ceramic material should not be subject to tensile stress.

- **Tightness of the heat exchanger**

The heat exchanger should be leak-tight because losses by leakage leads to lower efficiencies. Problems in this respect are to be expected at tube/tube or tube/tube-sheet joints.

- **Pulverised coal firing durability**

As the heat exchanger is charged with fly ash laden flue gas, it is susceptible to the hazards of corrosion and fouling.

The heat exchanger material must be resistant to flue gas and slag at high temperatures. Fouling of the heat exchanger has to be either prevented by appropriate upstream removal or minimised by deposit removal during operation. The decisive factors are the temperatures at which the heat exchanger is operated and the state of the arising ash or slag. In the temperature range above 1,400°C, i.e. higher than the ash fluid temperature, slag is removed in a molten state. At temperatures below the ash deformation point, the ability of soot-blowing to remove fouling deposits should be tested. At temperatures above the ash deformation point and below the ash fluid point, it may be necessary to heat up and melt the deposits.

Last but not least, there has to be safety during all operating states – start-up and shutdown and continuous operation, and cases of outages. None of these cases must lead to damage of the heat exchanger.

### 7.5.2.2 Selection of the Material

The material properties of the heat exchanger decide the upper limit of the process temperatures and hence the efficiency of the EFCC process. Figure 7.53 highlights the temperature-dependent stability of selected ceramic and metal materials. It is clear from the figure that, for the operation of the EFCC process in the high-temperature range, only ceramic materials can be considered (Kainer and Willmann 1987).

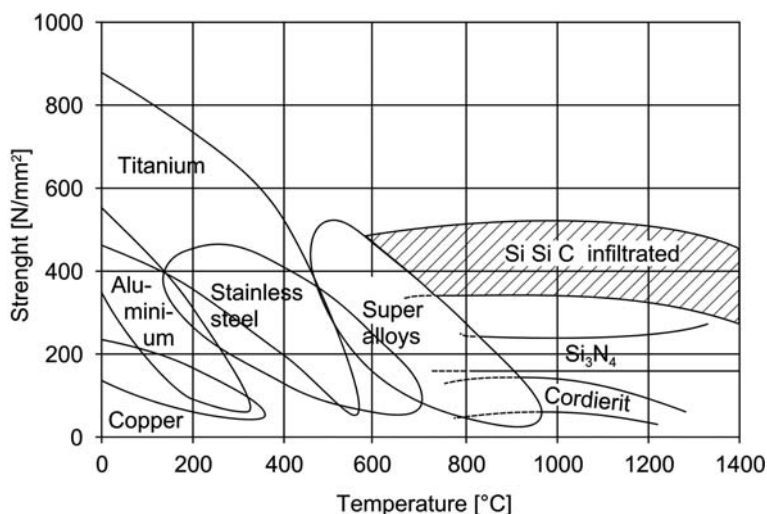


Fig. 7.53 Strength of metallic and ceramic materials (Kainer and Willmann 1987)

### Steels

Low-alloy steel types (represented typically by 15 Mo 3, 13 CrMo 4 4, 10 CrMo 9 10) can be used at temperatures of up to about 500°C. Higher-alloy ferritic steels – such as the type used for the final superheater stage in steam power plants, X 20 CrMoV 12 1 – or martensitic steels can be used up to temperatures of around 600°C. Austenitic steels can be used up to 750°C.

### Nickel-Based Alloys

Nickel-based alloys, where nickel is the main element of the alloy, have a higher temperature resistance and strength than austenitic steels. The temperature limit is about 850°C. Due to the composition, these materials are relatively expensive and rather difficult to handle.

### ODS Superalloys

ODS (oxide dispersion strengthened) alloys are powder metallurgical (PM) manufactured superalloys based on nickel or iron. They feature a high temperature resistance along with comparably good strength properties. Typical are Inconel MA 754 (Ni base) and PM 2000 (Fe base). They show highly promising properties such as high temperature and corrosion resistances up to temperatures of 1,150°C (Aquaro and Pieve 2007; Hurley et al. 2003).

### Ceramic Materials

Ceramic materials, with respect to temperature resistance paired with strength, are largely superior to metallic materials. Of disadvantage are the limited thermal shock

resistance and the limited tensile and bending strengths. Additional limitations to the use of ceramics arise due to their brittleness; these materials are not able to relax stress peaks by deformation. They are not at all ductile – on reaching the elastic limit fracture occurs immediately, without any deformation. Designs incorporating ceramics are suitable if they avoid high loads that are uncontrollable. Simple construction forms such as tubes and constructions that only put pressure stresses (rather than tensile) on the ceramic material are advantageous in this respect (Landfermann and Hausner 1988).

In principle, suitable materials for the high temperatures of EFCC heat exchangers are aluminium-based oxide ceramics and non-oxide ceramics such as  $\text{Si}_3\text{N}_4$  and SiC. The properties determining their suitability as construction materials for high-temperature heat exchangers are assessed in Table 7.13. According to this assessment, silicon carbide is the most suitable material meeting the requirements of the EFCC process. Table 7.14 draws a comparison of the properties of ceramic and other materials for the use in high-temperature heat exchangers. Fibre-strengthened ceramic materials are still under development. While they should be considered for use at high temperatures, problems persist with respect to their protection against long-term oxidation.

The corrosion resistance of ceramics is the major bottleneck. Only some types of ceramics, and only to a maximum of  $1,300^\circ\text{C}$ , meet the resistance requirements. This knowledge is the result of investigations into materials carried out in two coal-fired semi-industrial test plants within the framework of a research project (Kuhnle

**Table 7.13** Suitability of ceramic materials as construction materials for high-temperature heat exchangers (Baum 2001; Kuhnle et al. 1997; Fichtner 1992)

	Aluminium oxide	Silicon nitride $\text{Si}_3\text{N}_4$		Silicon carbide SiC	
	$\text{Al}_2\text{O}_3$	SSN RBSN	H(i)PSN	SSiC H(i)PSiC	SiSiC
Strength at high temperatures	+	(–)	+	+	(–) max $1,400^\circ\text{C}$
Thermal conductivity	(–)	(–)	(–)	+	+
Fatigue resistance	–	+	+	+	+
Resistance to oxidising atmospheres at high temperatures	+	–	(–)	+	+
Porosity/gas tightness	–	–	+	+	+
Resistance to molten coal slag			No resistance		

+ suitable, – not suitable, (–) only suitable to a limited extent *Silicon carbide SiC* (HPSiC hot pressed silicon carbide, HiPSiC hot isostatically pressed silicon carbide, SSiC sintered silicon carbide, SiSiC silicon-infiltrated silicon carbide)

*Silicon nitride  $\text{Si}_3\text{N}_4$*  (HPSN hot pressed silicon nitride, HiPSN hot isostatically pressed silicon nitride, SSN sintered silicon nitride, RBSN reaction-bonded silicon nitride)

**Table 7.14** Data for ceramic materials compared to other recuperator materials (Kainer 1988)

Material	Bulk density [g/cm <sup>3</sup> ]	Bending strength (20°C) [MPa]	Tensile strength [MPa]	Modulus of elasticity (20°C) [GPa]	Thermal			Open porosity [%]	Gas permeability [nPm]	Temp. limit [°C]
					expansion coefficient (20–1,000°C) 10 <sup>-6</sup> K <sup>-1</sup>	coefficient of thermal conductivity [W/mK]				
Alumina-rich fireclay	2.5	< 10	—	15	5	1–2		17	~ 1 · 10 <sup>-2</sup>	1,700
Cordierite	2.1	10–100	—	60	2	1–3		0–10	n.d.	1,200
Plasma-coated Al <sub>2</sub> O <sub>3</sub>	3.2–3.4	~ 20	—	22	3–8	1–3.5		10–12	~ 5 · 10 <sup>-3</sup>	1,200–1,500**
Silicon nitride	2.4	200	—	130	3	13		20	n.d.	1,350
Si-infiltrated SiC (SiSiC)	3.1	300	150	300	4.5	125–140		0	0	1,400
Sintered SiC (SSiC)	3.1	460	—	410	4.5	125–140		0	0	1,650
Stainless steel 25	7.9	—	900	210	11*	40		0	0	400
CrMo										
Tantalum	16.6	—	280	190	6.5*	55		0	0	700
Graphite	1.9	60	30	30	8*	120		0	0	200 (ox.) 2,500 (red)
Glass	2.23	70	—	68	3*	1		0	0	300

\* between 20–500°C

\*\* depending on posttreatment

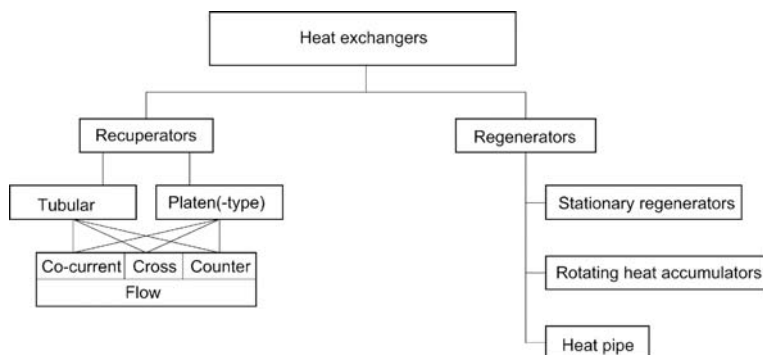
et al. 1997) and confirmed by experiments where materials were placed in an electrical furnace with synthetic flue gas and ash. Up to  $1,300^{\circ}\text{C}$ , the monolithic ceramic types – aluminium oxide, silicon nitride and silicon-infiltrated and sintered silicon carbide – showed promising corrosion behaviour for the hard coal used. In contrast, carbon fibre strengthened silicon carbide, with mechanical properties better suited to high-temperature heat transfer, showed poor oxidation behaviour despite the coating (Baum 2001).

When the materials in the preceding paragraph were tested in a cyclone slag-tap furnace at high flue gas temperatures of  $1,500^{\circ}\text{C}$ , all showed severe damage even after less than 30 h, which excludes their application in high-temperature heat transfer components. The corrosive attack comes especially from the aggressive molten slag. The use of ceramic materials under these conditions needs further development. Apart from finding suitable ceramic materials and construction types for them, development of how to make adequate connections and joins of ceramic constructions is necessary. For temperatures below  $1,150^{\circ}\text{C}$ , the oxide dispersion strengthened (ODS) superalloy PM 2000 seems to be suitable for use. This metallic material, in comparison to monolithic ceramics, has great mechanical property and manufacturability advantages.

### 7.5.2.3 Classification of Heat Exchangers

There are primarily two techniques of heat transfer available: recuperative heat exchangers (or recuperators) and regenerators. While recuperators transfer heat from one heat-carrying medium to the other via separating walls, regenerators exchange heat after a time lag via intermediary media. Figure 7.54 gives an overview of heat exchanger technologies (Kainer and Willmann 1987). Recuperators work continuously in transferring heat, while regenerators, by alternating between heat storage and heat dissipation, are discontinuous processes.

Only regenerators with many single-storage elements, which, as they circulate, alternate the processes of heat storage and heat dissipation, make quasi-continuous operation possible. Regenerators with temperature-resistant, thermally conductive

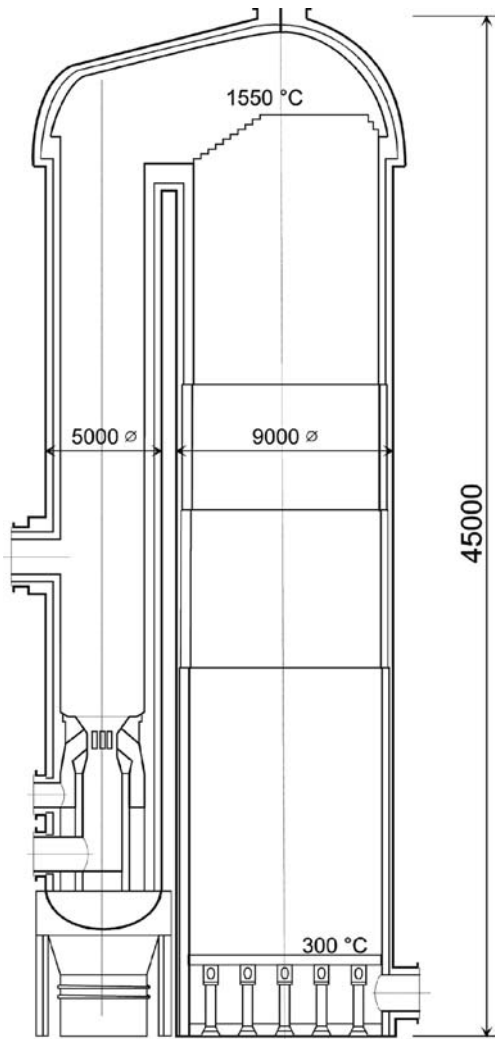


**Fig. 7.54** Heat exchanger systems (Kainer 1988)



storage elements such as ceramics, as well as temperature-resistant supports, allow high working temperatures. Disadvantages of regenerators are gas exchanges or gas losses associated with every switching and the extra cost of construction and control engineering (Kainer 1988).

A well-known example of regenerators is hot blast stove for air preheating in blast furnace processes (see Fig. 7.55). Part of the hot blast stove is a static heat accumulator which is built of single ceramic hollow blocks placed upon each other and penetrated by the air flow. The heat accumulator is placed on a metal grid connected to a furnace which, in the case pictured, is mounted externally. The heat accumulator and furnace are surrounded by thermal insulation and steel plating. A



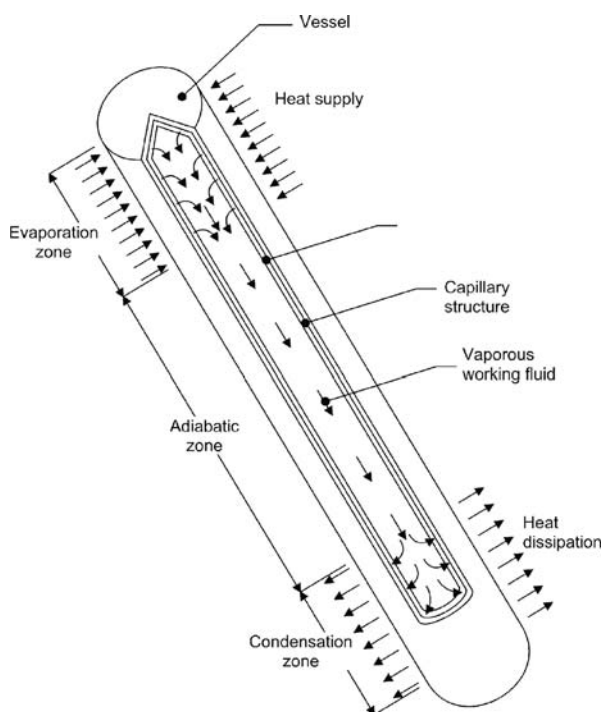
**Fig. 7.55** A typical regenerator – a hot blast stove with an external furnace for blast furnace operation (Kainer 1988)

complete air preheating plant for a blast furnace needs at least two, often up to four, regenerators because the single regenerator can only be operated discontinuously. By means of such plants, it is possible to heat air flowrates of up to 450,000 Nm<sup>3</sup>/h to temperature maxima of 1,400°C at pressures of up to 6 bar (Kainer 1988).

Another heat exchanger type considered for EFCC processes is the regenerative Ljungstroem heat exchanger, featuring a rotating heat accumulator (Wilson et al. 1991; Wilson 1993a). However, further development of sealing engineering is necessary to reduce the leakage loss that results from rotation. One possibility is to rotate the cylindrical heat accumulator only in short intervals. The gaskets are then released and lifted only during rotation, otherwise pressed against the heat accumulator. Another technology considered is regenerator where rectangular heat-accumulating blocks circulate between the hot and the cold gas flow. This way the lengths to be sealed are shorter than for a rotating heat exchanger (Wilson 1993b).

The biggest problem with all regenerators, however, is fouling. Because the medium which temporarily stores the heat is in contact with both the polluted flue gas and the clean working fluid of the gas turbine, valves and gaskets are subject to fouling. This way, fly ash or alkalis can also pollute the turbine working fluid.

Yet another technology considered for high-temperature processes is heat pipe, the principle of which is shown in Fig. 7.56 (Bliem 1985). It is a closed pipe, one



**Fig. 7.56** Schematic drawing of a heat pipe (from Bliem 1985, © 1985, with permission from Noyes Publications)

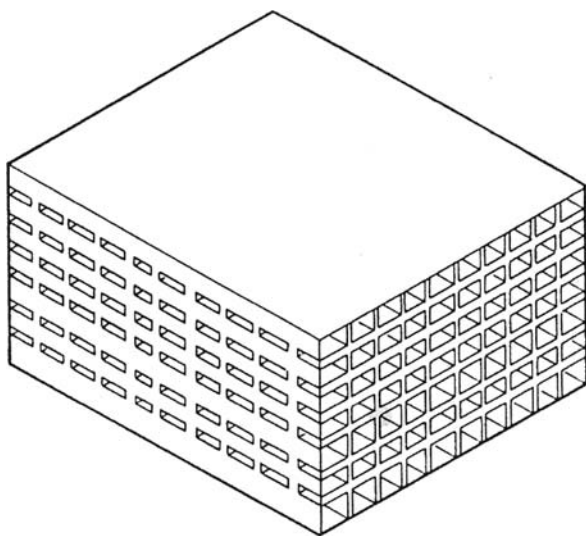
half of which projects into the untreated flue gas, the other into the cleaned gas. The tube, the inside of which is in a vacuum, has a capillary structure saturated with a liquid. When the half of the tube in the flue gas is heated, the working fluid vaporises and flows to the other end of the tube where, dissipating heat, it condenses internally. The capillary action transports the formed condensate back to the heating zone.

For use in an EFCC process, the heat carrier and its supporting structure have to be adjusted to high temperatures. For temperatures up to  $1,600^{\circ}\text{C}$ , lithium is considered the most suitable heat carrier and SiC the most suitable structural material (Groll 1980). These heat pipes, however, are not state of the art. As with other types of heat exchangers, heat tubes require a solution to the problem of corrosion. Heat pipes with sodium as the working fluid, transferring heat between two small-scale fluidised beds at  $800^{\circ}\text{C}$ , have been successfully tested (Kuhn 2007).

Recuperators that are used at high temperatures have to be made of ceramic materials. Up until now, there have been two different types of designs developed: the modular or block system and the shell-and-tube construction.

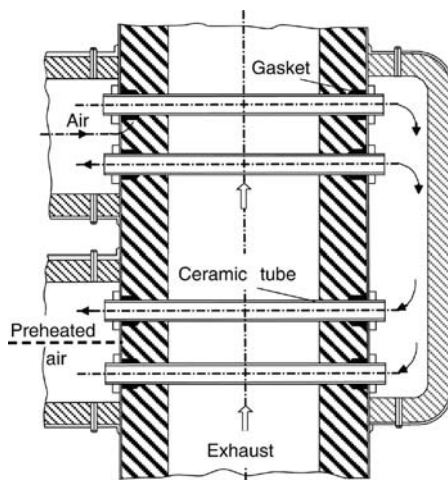
Figure 7.57 shows the block of a module-type heat exchanger. By series connection of the single modules it is possible to conduct mass in a cross-counterflow regime. The modules consist of extruded plates which are stacked together and joined by a special substance during baking. Their susceptibility to fouling and clogging and their limited accessibility for cleaning suggest that such module-type heat exchangers are less suitable for a coal-fired EFCC process.

Figure 7.58 shows the working principle of a ceramic cross-flow recuperator. The straight ceramic tubes are flexibly mounted at both ends so that the tubes can extend without stress. Plugging gaskets of ceramic fibres serve to minimise leakages. Another possibility is to clamp the tubes at one end – preferably at the colder



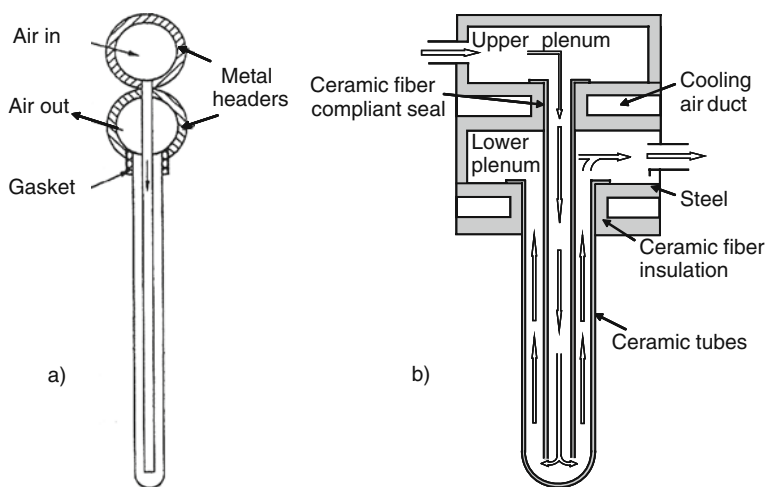
**Fig. 7.57** Unit of a module-type heat exchanger (from Bliem 1985, © 1985, with permission from Noyes Publications)

**Fig. 7.58** Working principle of a ceramic recuperator (Kainer and Willmann 1987)



one – so as to mount them flexibly while allowing for extension at one end (Kainer and Willmann 1987).

The principle of clamping at one end is also applied in the construction of the double-tube recuperator shown in Fig. 7.59. Both tubes are clamped at one end and can freely extend in the other direction. The tubes are suspended vertically so gravity affects the gasket between the housing and the tube flange. The outer tube has a diameter of about 95 mm, a wall thickness of 6 mm and a length of nearly 2,000 mm. The tube is made of SiC (Bliem 1985; Harkins and Ward 1989).



**Fig. 7.59** Tube-in-tube recuperators (b from Bliem 1985), © 1985, with permission of Noyes Publications)

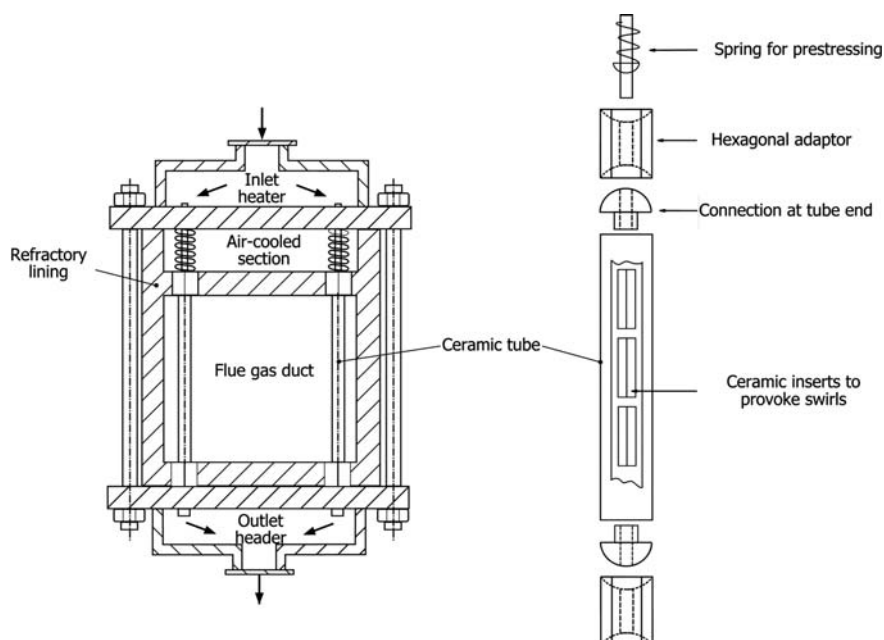


Fig. 7.60 Recuperator by Hague International (LaHaye 1989, 1986)

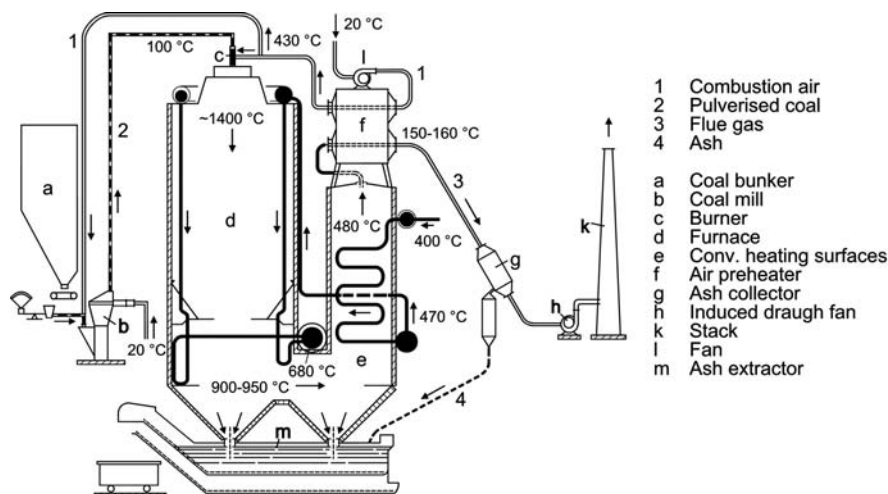
When ceramic heat exchangers are used in an open process with air as the working fluid of the gas turbine, attention must be paid to the pressure difference between the air and the flue gas. Given that ceramic materials, with their minimal ductility, should preferably be loaded with compressive stress, it is logical to conduct the pressure-free flue gas inside and the working fluid outside the tubes of a shell-and-tube heat exchanger. Conducting the slag- or ash-carrying flue gas inside the tubes is a valid counterargument, though.

The heat exchanger concept for EFCC pursued by Hague International is to conduct the clean working fluid of the gas turbine inside the tubes and the hot flue gas outside, as shown in Fig. 7.60. The tubes are pre-stressed by means of a system of springs in order to be able to load the ceramic material with compressive stress and to seal the tubes. Tube circumferences typically measure between 75 and 100 mm and have a length of about 1,200 mm. Semi-circular fittings and adaptors guarantee the absorption of shearing forces (lateral extensions) (LaHaye and Feldmann 1986; LaHaye and Zabolotny 1989; Vandervort 1991). A heat exchanger designed for a demonstration power plant is described in Sect. 7.5.3.2.

### 7.5.3 State of Development

Although the EFCC process has been well known for a long time, only the further development of ceramic materials during the past 10–15 years has given it a new





**Fig. 7.62** Schematic diagram of the EFCC plant in Ravensburg, Baden-Württemberg (Keller and Gaehler 1961)

The schematic diagram of the combustion plant is shown in Fig. 7.62. The furnace was refractory-lined and designed as a two-pass construction. The pulverised coal burner was mounted at the furnace top. The air, preheated to  $450^{\circ}\text{C}$  in the recuperator, was heated by convective heating surfaces arranged in the second pass and then by radiant wall heating surfaces. The heating surfaces were designed as in conventional steam generators, with inlet headers, single tubes and outlet headers. The tube dimensions were an inside diameter of 32 mm and a wall thickness of 3 mm; the number of single tubes of the  $6.6\text{ MW}_{\text{el}}$  plant in Coburg was 320 (Keller and Gaehler 1961). At a gas turbine inlet temperature of  $710^{\circ}\text{C}$ , the highest tube wall temperatures were between  $770$  and  $790^{\circ}\text{C}$ , which austenitic materials could still cope with. The highest alloyed material used was austenite of the 16 Cr13Ni type. All plants built in Germany reached 120,000 h of operation; only the plant in Coburg was in service for more than 160,000 h (by 1986) (Bammert 1986).

Another type of EFCC process using metal as a construction material was put into practice in a model power plant in Völklingen, Saarland (Germany). In it, the in-bed heat transfer surfaces of two fluidised bed modules were used to preheat the air to about  $700^{\circ}\text{C}$ . The gas turbine inlet temperature could be raised to  $820^{\circ}\text{C}$  by additionally firing a gaseous fuel. The combustion gases of both fluidised beds were conducted into the pulverised coal firing of the steam generator, while the gas turbine exhaust was made use of as a fluidising medium and oxygen carrier in both fluidised bed modules and as secondary air for the pulverised coal firing. Topping the gas turbine made the efficiency rise to 2% higher than the steam process (Stoll and Bleif 1986).

The programme “Combustion 2000”, funded by the US Department of Energy (DoE) for the development of efficient technologies for the generation of power from coal, also pursued the EFCC process as a concept. Contracts were issued to two

independent consortia, led by the Foster Wheeler Development Company (FWDC) and United Technologies Research Center (UTRC), respectively. The development of the overall process, called “High Performance Power Systems” (HIPPS), however, only relied on currently available technology. The most recent feasible concept design involved a metallic heat exchanger which is used to preheat air, with natural gas additionally being fed to raise the temperature.

Besides the high-temperature heat exchanger, the Foster Wheeler concept design included a pyrolyser and a char-fired combustion system, with heat being transferred to preheat the air and to produce steam. Air is preheated up to 760°C using tube banks constructed of alloyed steel; in a topping combustor that is fired with fuel gas from the pyrolyser, air is further heated to a gas turbine inlet temperature of 1,288°C.

The UTRC HIPPS concept was based on a turbine working fluid heat exchanger outlet temperature of 1,000°C and a gas turbine inlet temperature of 1,260°C. With natural gas contributing about one third of the thermal power output, an efficiency of 50.7% (LHV) was given (Klara 1994a, 1994b; Ruth 1997, 2001). However, Phase III of the HIPPS programme, which would have involved construction of a demonstration plant, was terminated and a demonstration plant will not be built (Benson 2000).

### 7.5.3.2 EFCC Processes with Ceramic Heat Exchangers

In the 1970s and 1980s, several US companies carried out investigations into the use of ceramic heat exchangers in EFCC processes. The results, however, have not been translated into practice in commercial EFCC plants.

In 1977, Solar Turbines Inc., supported by the US Department of Energy and the Electric Power Research Institute (EPRI), started a project investigating a ceramic high-temperature heat exchanger (SolarTurbines 1980). In the course of this research project, material tests were carried out in different atmospheres and at different temperatures, followed by a strength test. Furthermore, investigations into joining techniques of ceramic heat exchanger components were a subject of the research. In the end, a vertical shell-and-tube heat exchanger with a counterflow configuration made of silicon carbide tubes was built.

In a subsequent project, this heat exchanger was successfully subjected to 1,370°C hot flue gas from the firing of oil (Ward et al. 1983). There are no subsequent publications on tests using coal flue gas, which suggests that operational problems arose.

In the area of indirectly fired gas turbine processes, Solar Turbines further planned to use a ceramic high-temperature heat exchanger in waste incineration. The research project was to conclude with field tests using a “high-pressure ceramic heat exchange system” (HiPHES) with a thermal capacity of 10 MW in a waste incineration plant in Houston, Texas (Harkins and Ward 1989). Again, there are no current publications on their experiences.

From the mid to the end of the 1970s, investigations into high-temperature heat exchangers in coal flue gas atmospheres were carried out by AiResearch with the support of the Electric Power Research Institute. For instance, a small recuperator



model was tested at a flue gas temperature of  $1,260^{\circ}\text{C}$ . The material temperature in this test was  $1,093^{\circ}\text{C}$  (Pietsch 1978). In a following project, the fundamentals of the manufacturing process and the design of the heat exchanger were adapted to a closed gas turbine process using argon as the working fluid of the gas turbine. Use was also made of the previous material characterisations during this adaptation (Coombs et al. 1979).

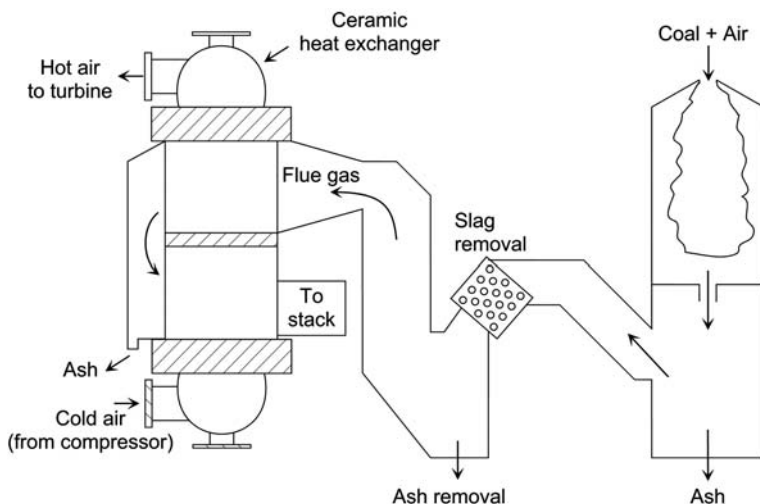
Probably the most advanced development programme of an EFCC process incorporating a ceramic heat exchanger was carried out in the USA by Hague International and other industrial partners and supported by the US Department of Energy. Within the framework of the first phase, from 1987 to 1989, suitable materials were tested; high-pressure and high-temperature experiments were carried out with different heat exchanger tubes; the effect on fouling and corrosion by the coal flue gases was determined; and methods of cleaning were trialled (Vandervort and Orozco 1992; LaHaye et al. 1990). According to the published data, these investigations show that it is possible to control the material stress, fouling and slagging at material temperatures of  $1,200^{\circ}\text{C}$ .

In Phase II of the "Combustion 2000" programme (1990–1994), an experimental plant with a thermal capacity of 7.4 MW was built, equipped with a ceramic heat exchanger of 2 MW. The execution of the project was halted in 1996, however, because the operation of the plant was impossible beyond a period of 50 h (DoE 1997). Phase III, which was designed to retrofit an existing coal-fired power plant to be an EFCC process, and had already begun in 1994, was stopped as well.

The design of the Phase III demonstration retrofitted power plant, Warren, had an efficiency of 37%, but was stopped before it was completed. The gross gas turbine output was  $22\text{ MW}_{\text{el}}$  and the gross steam turbine output reached  $48\text{ MW}_{\text{el}}$  – so the resulting total output, taking into account an auxiliary power requirement of  $4\text{ MW}_{\text{el}}$ , amounted to  $66\text{ MW}_{\text{el}}$  (LaHaye and Bary 1994). The planned outlet temperatures of the high-temperature heat exchanger were at roughly  $900^{\circ}\text{C}$  at tube wall temperatures of about  $1,150^{\circ}\text{C}$ . The layout seems to be such that tube wall temperatures should be kept well below  $1,200^{\circ}\text{C}$ . Since the ceramic tubes used are supposed to be suitable for temperatures up to  $1,480^{\circ}\text{C}$ , this limitation to  $1,200^{\circ}\text{C}$  could be an indication of fouling and corrosion problems. The low efficiency of the planned retrofit is to be put down to the low gas turbine inlet temperature and the applied steam cycle ( $470^{\circ}\text{C}$ , 60 bar) being not of high enough quality because of the retrofitting.

A schematic diagram of the furnace and heat exchanger of the  $7.4\text{ MW}_{\text{th}}$  EFCC test plant is shown in Fig. 7.63. The planned design of the Warren power plant described hereafter follows the layout of the test plant. The combustion takes place in a slag-tap furnace, the operation of which is air-staged for  $\text{NO}_x$  reduction. The furnace was designed as a membrane wall construction with water/steam cooling, refractory-lined to minimise the heat absorption by the walls.

Particle-laden gases exit the combustor and enter the slag screen to remove particles above a certain size. This should prevent ash deposition in the ceramic heat exchanger. The slag screen was to consist of single rods and to be designed such that particles larger than  $12\text{ }\mu\text{m}$  would not follow the flue gas as it deflected but collect on the rods.

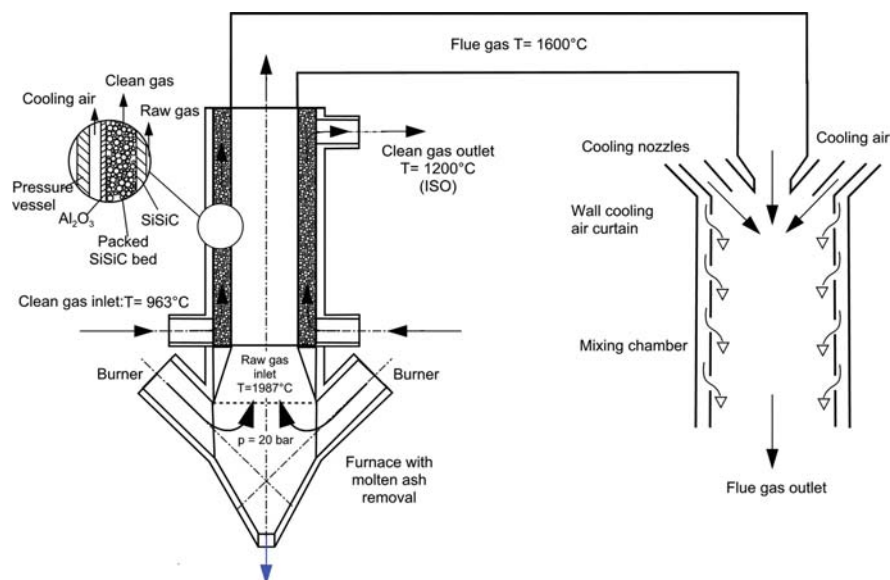


**Fig. 7.63** Schematic diagram of a 7.4 MW<sub>th</sub> EFCC test plant (Vandervort 1991, Vandervort and Orozco 1992)

The dimensions of the ceramic heat exchanger of the Warren power plant were to be  $26.8 \times 8.2 \times 2.4$  m, comprising 700 tubes in total. The flue gas was to flow on the outside of the tubes through four heat exchanger passes altogether, the compressed air through the vertical tubes. The ceramic tubes of one pass were to have a length of 4.9 m and an outside diameter of 10 cm. The heat exchanger material to be used was ceramic on an aluminium/silica carbide base (LaHaye and Bary 1994). The pressure loss on the side of the flue gas given for the dust collector and the heat exchanger was below 50 mbar, and on the side of the air in the heat exchanger, lower than 0.2 bar. The total leakage loss was assumed to be less than 0.5% (LaHaye and Bary 1994).

In Germany, in the context of the development of pressurised pulverised coal fired furnaces, the use of a high-temperature heat exchanger was also considered (Hannes 1986). In contrast to the variant shown in Fig. 7.47, the concept design included a low-temperature heat exchanger as well as a high-temperature one. After the transfer of the flue gas heat to the cleaned gas, the flue gas, at a temperature still above the ash fluid temperature, was to be cooled below the ash deformation temperature by adding warm air at 300°C in order to prevent uncontrollable sticky deposits in the following convective heat exchanger. The air guidance was also chosen to prevent sticking in the multi-fuel combustion chamber by producing an air curtain (Neumann et al. 1996).

The firing system is arranged as a cyclone furnace boiler, where part of the ash is removed in a molten state. In the radiation section which follows, heat is transferred by a high-temperature heat exchanger. The construction of the heat exchanger can be seen in Fig. 7.64. It consists of two concentric ceramic tubes set in an air-cooled pressure vessel. The flue gas transfers its heat by radiation to the ceramic tube which forms the furnace wall. Within the cross-section of the two ceramic tubes, the



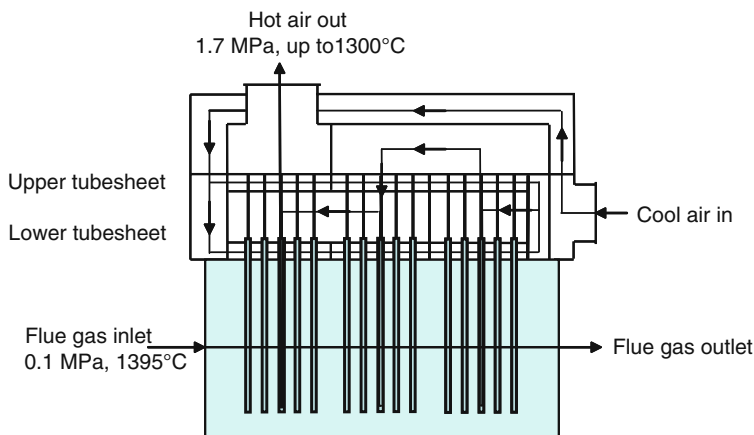
**Fig. 7.64** An EFCC process with a furnace, heat exchanger and multi-fuel combustion chamber (Neumann et al. 1996)

working fluid of the gas turbine absorbs the dissipated heat, mostly by convection. The annular clearance contains ceramic packing to improve the heat transfer. The exterior pressure vessel is cooled by conducting air between the exterior ceramic tube and the vessel wall, so that the ceramic heat exchanger is not loaded with pressure (because both the flue gas and the working fluid of the turbine are pressurised).

A  $600\text{ MW}_{\text{el}}$  plant needs eight parallel trains each with a furnace and a high-temperature heat exchanger, where the high-temperature heat exchanger has a height of 12.3 m and the pressure vessel a diameter of 3.5 m. The efficiency depends on the temperature of the gas cleaning stage. With flue gas cleaning at the cold end, the efficiency is 48%; with hot gas cleaning at  $850^{\circ}\text{C}$ , the calculated efficiency amounts to 51% (Neumann et al. 1996).

The Italian company Ansaldo Ricerche, in collaboration with its European partners, is undertaking a programme which aims to realise a ceramic heat exchanger module. Initial programmes concentrated on the development of production techniques and material development for reliable and cost-effective ceramic tubes for use in aggressive and high-temperature environments. Research has mainly concentrated on coated carbon fibre reinforced silicon carbide and one monolithic SiC. Continuation of this research focussed on the design of a high-temperature heat exchanger module.

The design of the heat exchanger and its modules is shown in Fig. 7.65. The heat exchanger is designed to operate with the flue gas at  $1,395^{\circ}\text{C}$  and 0.1 MPa and the compressed air at 1.7 MPa with a maximum temperature of  $1,300^{\circ}\text{C}$ . The flue gas and compressed air flow in opposite directions through a series of modules.



**Fig. 7.65** Ceramic heat exchanger module (Benson 2000)

Those modules which are only exposed to temperatures below 850–900°C can be constructed from heat exchange tubes made from ODS alloys. The tubes in the high-temperature modules are ceramic. The structural elements of the heat exchanger are made from metallic materials and cooled by air. After preheating, the air is conducted through the ceramic tubes, which are suspended in the flue gas, where the air is heated to the TIT of 1,300°C. The flue gas flows directly through the module, transferring heat to the compressed air.

A ceramic heat exchanger is being tested at a 5 MW multi-fuel boiler burning coal to generate the flue gas. In the proposed design, the heat exchanger consists of four modules: two modules containing 42 ceramic U-tube arrangements and two modules containing 40 ODS alloy U-tubes. The outer tube dimensions are 1.55 m in length by 7.5 cm outer diameter. The modules are spaced 50 cm apart, with soot-blowing equipment incorporated between them. It is planned to initially use monolithic SiC tubes in the ceramic heat exchanger modules and then swap to CMC (ceramic matrix composite) tubes based on C/C-SiC when a suitably developed material becomes available (Benson 2000).

### 7.5.4 Conclusions

The EFCC process offers the advantage of high efficiency; however, the problems associated with the high temperatures are not yet solved. The major bottleneck seems to be the corrosion attack by liquid slag when the ceramic heat exchangers are exposed to temperatures of 1,400–1,600°C. At temperatures up to 1,300°C, oxide dispersion strengthened alloys are preferable as they are easier to handle. Whereas for large-scale coal-fired power stations the technology is not yet applicable, the technology could be suitable for smaller biomass-based systems with lower gas turbine inlet temperatures.

## 7.6 Integrated Gasification Combined Cycle (IGCC)

### 7.6.1 History of Coal Gasification

Gasification, in the broadest sense, means the transformation of a solid carbonaceous fuel into a gas with a useful calorific value. The wider variety of applications and the advantages of a gaseous fuel in contrast to solid coal spurred the development of coal gasification. This development began in the 19th century with the powering of street lighting by coal gas (1812 saw the first commercial gasworks for illuminating gas production in London), while in later years gasification for heating purposes became the dominant motivator. This gas, called town gas, was produced in a discontinuous pyrolysis process similar to a coking process. From about 1880, this method was superseded by the water gas shift reaction, which was then used for town gas production until the middle of the 20th century. In this process, with the supply of heat and steam, the solid carbon is transferred into a mixture of carbon monoxide and hydrogen, as expressed in Eq. (7.37). The rising availability of cheap natural gas put an end to the consumption of town gas in Europe from about 1970, and coal gasification became limited to niche applications (Higman and van der Burgt 2008).

A step of great importance in the development of gasification was the commercialisation of cryogenic air separation by Carl von Linde in the 1920s, which then made it possible to operate a continuous, oxygen-blown gasification process for the production of synthesis gas (commonly known as syngas) and hydrogen. Developments took place around that period that were the precursors of today's gasification technologies: the Winkler process (a fluidised bed gasifier process) in 1926, the Lurgi process (a fixed bed coal gasification process) in 1931 and the Koppers-Totzek process (an entrained-flow process for the gasification of pulverised coal) in 1940. After these processes had been established, there were only minor advances in the following 40 years, save for the rise of a petrochemical industry based on coal gasification and the Fischer – Tropsch synthesis process by the Sasol Company in South Africa.

In the wake of the oil crisis in the 1970s and the feared shortage of natural gas and oil, extensive efforts were undertaken then and in the 1980s to further develop and demonstrate gasification technologies for the production of syngas, liquid or gaseous energy sources, or electric power. By way of example, Lurgi and British Gas developed a molten bath gasifier; Koppers and Shell together worked on a pressurised version of the Koppers-Totzek gasifier; and Rheinbraun developed the high-temperature Winkler process. As oil prices sunk, however, the interest in coal gasification or liquefaction decreased at the end of the 1980s, restricting further development.

Coal gasification technology for electric power production was demonstrated in Europe in several large-scale plants (Lünen, 170 MW, 1972; Buggenum, 250 MW, 1992; Puertollano, 335 MW, 1997) with success. The motivation for pursuing this technology in the future is the potential for better environmental performance at a lower marginal cost. A broad introduction into the electricity generation market of

this technology, however, has not yet taken place due to its cost disadvantages and low availability.

Nonetheless, there are a great number of gasification plants worldwide at the moment. The cumulative gasification capacity reported for 2008 ranged around 70 GW<sub>th</sub> (Higman and van der Burgt 2008). These plants are mostly designed and built to produce syngas for ammonia, hydrogen or transportation fuel production. The feedstocks in many cases are difficult fuels, such as refinery waste or petrol coke, which are hard to exploit with other technologies. Here, the advantages of the low environmental impact of gasification come fully to fruition.

A strong increase over the past 10 years in installed gasification capacities is noticeable. The various advantages of gasification technology give reason to believe that gasification will see a revival:

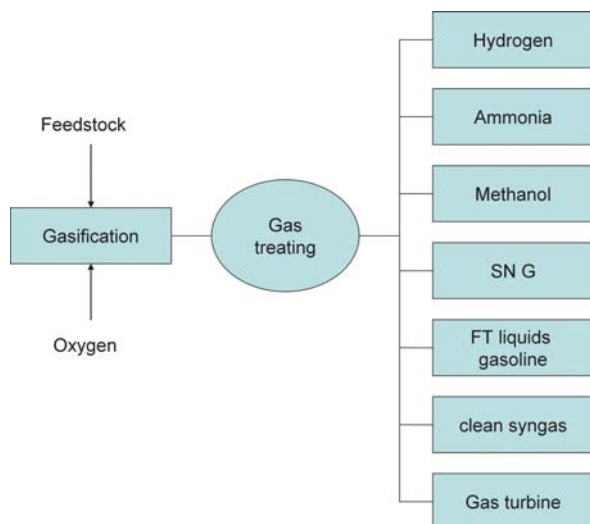
- Gasifiers are suitable for a wide range of fuels. In an entrained-flow gasifier for instance, coal can be used together with biomass and residual material. Besides solid fuels, liquid fuels can also be used.
- The emissions of all gaseous pollutants and trace components, given that gas cleaning has to be installed for process-engineering reasons, are significantly lower than from conventional power plants.
- Using two additional common process steps (a CO shift reactor and CO<sub>2</sub> capture), it is possible to separate CO<sub>2</sub>. The required additional effort is lower, owing to the process pressure and the small volumetric flow, than in downstream CO<sub>2</sub> scrubbing following a steam power cycle. Considering this, the advantage in efficiency that gasification already has would become still greater. The present disadvantage of higher capital costs could be balanced out.
- Gasification technology offers the highest product flexibility. Gasification together with synthesis gas generation form the first process step to produce liquid fuels (Fischer – Tropsch), gaseous secondary energy sources such as hydrogen or synthetic natural gas, and methanol or ammonia for the chemical industry (Higman and van der Burgt 2008).

## ***7.6.2 Applications of Gasification Technology***

### **7.6.2.1 Generation of Secondary Energy Sources**

Gasification technologies are suited to providing a high-energy gas for the production of basic products for the chemical industry or of secondary energy sources. Figure 7.66 presents an overview of the various possible uses of product gases from gasification processes.

The synthesis process for the generation of the final product determines the entire cycle and defines the requirements for the gasification process and the gas treatment. The major gas components and allowable gaseous pollutants have to be taken into consideration. The requirements differ from process to process but, in general, the level to be met is high. The sulphur content by volume, for example, has to be



**Fig. 7.66** Production possibilities with gasification

below 1.0 ppm or even below 0.1 ppm. A summary of the gas purity requirements of different synthesis processes can be found in Radtke et al. (2006), Ogriseck and Meyer (2005) and Higman and van der Burgt (2008).

It is plausible to say that for all processes, nitrogen is an unwelcome component in the final gasification gas. Given that removing nitrogen from the gas produced is more complex than the fractionation of air, almost all industrial-scale gasification processes are operated with oxygen. In principle, alternative methods to an oxygen-blown gasification process exist, such as allothermal processes, where heat is transferred from external sources, thereby generating a nitrogen-free gas product. These processes, which are considered for medium-scale biomass applications (see Sect. 6.3), are significantly more costly for larger-scale units and are therefore not today's generally accepted practice. Instead, high-temperature entrained-flow gasification is the technology considered feasible for the production of basic chemical substances or secondary energy media. In this process syngas is produced, which consists almost exclusively of  $H_2$  and CO. Further processing of the gas, such as the CO shift process, is then required so that it conforms to compositional requirements as a product. The removal of trace elements such as sulphur, phosphorus and chlorine is required so that deactivation of the catalyst of the syngas is minimised.

### 7.6.2.2 IGCC With and Without $CO_2$ Capture

The integrated gasification combined cycle (IGCC) produces electricity from a solid or a liquid fuel. First, the fuel is converted to a syngas, which is then converted to electricity in a combined cycle power plant consisting of a gas turbine process and a steam turbine process with a heat recovery steam generator (HRSG). The combined

cycle technology is similar to the technology used in modern natural gas fired power plants.

### IGCC Without CO<sub>2</sub> Capture

Figure 7.67 shows the principle units of a coal-based IGCC plant without CO<sub>2</sub> capture. The coal is supplied to the gasifier, where it is partially oxidised under pressure (30–80 bar). The plant uses oxygen as the oxidant and therefore has an air separation unit (ASU). In the gasifier, which is normally of the entrained-flow slagging type, the temperature may exceed 1,500°C. In addition to its chemical energy (heating value), the hot raw syngas contains sensible heat, which may be recovered in heat exchangers to produce steam for the steam turbine. It would be desirable to clean the raw syngas without cooling and to deliver the hot syngas to the gas turbine, which would result in a higher efficiency. However, hot gas cleaning (discussed in Sect. 7.6.5.6) is not state of the art, and proven technologies operate at near ambient temperatures. In the gas clean-up process, particles, sulphur and other impurities are removed.

For the sake of efficiency, it is practical to integrate the air separation as completely as possible into the IGCC process. It is talked of as 100% integration if the air flow compressed in the compressor of the gas turbine is fed entirely to the air separation unit and the nitrogen stream separated under pressure is completely

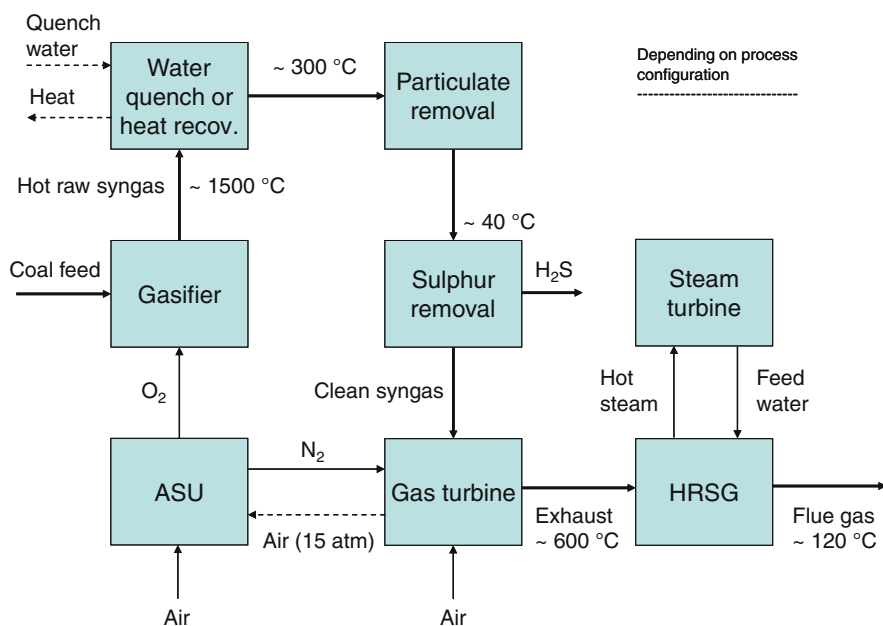


Fig. 7.67 An IGCC process without CO<sub>2</sub> capture (Maurstad 2005)



expanded in the gas turbine. Full integration yields the highest efficiency; partial integration can result in a higher output and a higher operational flexibility, for example in the start-up process.

### IGCC with CO<sub>2</sub> Capture

The IGCC power plant offers favourable conditions for capturing CO<sub>2</sub>, because the separation of CO<sub>2</sub> under pressure involves less energy loss than downstream removal from the atmospheric flue gas. The capture process of CO<sub>2</sub> in an IGCC power plant needs two additional components, as shown in Fig. 7.68:

- a so-called shift reactor, which converts the CO of the syngas into CO<sub>2</sub> and hydrogen using water vapour, and
- a unit for CO<sub>2</sub> removal by chemical or physical absorption, which removes the CO<sub>2</sub> from the hydrogen/carbon dioxide mixture.

The gas turbine is thus fed with hydrogen or with a fuel rich in hydrogen.

The two additionally necessary process steps are discussed in more detail in Sects. 7.6.5.4 and 7.6.5.5.

In an IGCC process with CO<sub>2</sub> capture the efficiency is lower because, due to the exothermic shift reaction, some of the chemical energy of the fuel is converted into

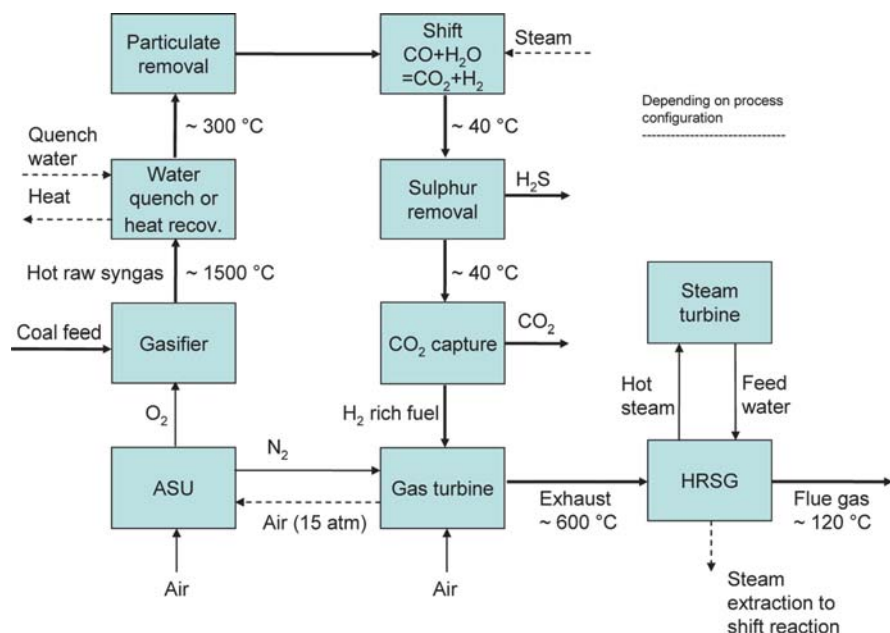


Fig. 7.68 IGCC process with CO<sub>2</sub> capture (Maurstad 2005)

heat at a low temperature, and this heat energy can only be converted to electrical power with a lower efficiency than chemical energy. In most gasification processes, the steam – carbon ratio is too low for the shift reaction, so high-quality steam has to be added. In addition, energy is needed for compressing the  $\text{CO}_2$ . On top of that, energy is also needed for the separation process, for example for regenerating a solvent.

### 7.6.2.3 Factors Affecting the Efficiency of an IGCC

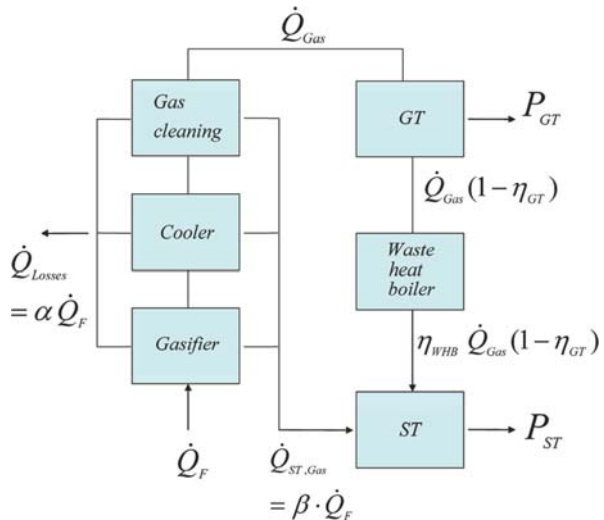
Figure 7.69 shows the energy flows of a simplified IGCC system. In the gasification stage, the chemically bound energy of the fuel  $\dot{Q}_F$  is converted into the energy  $\dot{Q}_{\text{Gas}}$  of the syngas in the gasification island (comprised of the gasifier, syngas cooling and gas cleaning stages).  $\dot{Q}_{\text{Gas}}$  includes both the sensible heat and the chemically bound energy.

The (warm gas) gasification efficiency,  $\eta_{\text{Gas}}$ , is defined as

$$\eta_{\text{Gas}} = \frac{\dot{Q}_{\text{Gas, Chem.}} + \dot{Q}_{\text{Gas, sensible}}}{\dot{Q}_F} \quad (7.14)$$

The gasification efficiency of the gasification island is dependent on the heat losses  $\dot{Q}_{\text{Loss}}$  and the steam production  $\dot{Q}_{\text{St, Gas}}$ , which mainly arises from syngas cooling. The dimensionless parameters  $\alpha$  and  $\beta$  relate the losses and the steam production to the fuel input:

$$\alpha = \frac{\dot{Q}_{\text{Loss}}}{\dot{Q}_F} \quad \beta = \frac{\dot{Q}_{\text{St, Gas}}}{\dot{Q}_F} \quad (7.15)$$



**Fig. 7.69** A simplified IGCC process for efficiency calculations

whereas the cold gas efficiency,  $\eta_{\text{Cold Gas}}$

$$\eta_{\text{Cold Gas}} = \frac{\dot{Q}_{\text{Gas, Chem}}}{\dot{Q}_F} \quad (7.16)$$

only considers the chemically bound energy of the product gas.

In an IGCC system, power is produced in gas and steam turbines. The efficiency  $\eta_{\text{IGCC}}$  is defined as

$$\eta_{\text{IGCC}} = \frac{P_{\text{ST}} + P_{\text{GT}}}{\dot{Q}_F} \quad (7.17)$$

In the gas turbine, the energy  $\dot{Q}_{\text{Gas}}$  is converted into power  $P_{\text{GT}}$  with the efficiency  $\eta_{\text{GT}}$ :

$$P_{\text{GT}} = \eta_{\text{GT}} \dot{Q}_{\text{Gas}} = \eta_{\text{GT}} \cdot \eta_{\text{Gas}} \cdot \dot{Q}_F \quad (7.18)$$

Steam is produced both in the gasification island  $\dot{Q}_{\text{St, Gas}}$  and in the waste heat boiler  $\dot{Q}_{\text{St, WHB}}$ .

The heat which is not converted into power in the gas turbine is fed to the waste heat boiler and converted with the efficiency of the waste heat boiler  $\eta_{\text{WHB}}$  into steam:

$$\dot{Q}_{\text{St, WHB}} = \eta_{\text{WHB}} \cdot \dot{Q}_{\text{Gas}} \cdot (1 - \eta_{\text{GT}}) = \eta_{\text{WHB}} \cdot \eta_{\text{Gas}} (1 - \eta_{\text{GT}}) \cdot \dot{Q}_F \quad (7.19)$$

Both the steam of the waste heat steam generator and the steam from the gasification island are converted into power with the efficiency  $\eta_{\text{ST}}$  via steam in the steam process:

$$P_{\text{ST}} = \eta_{\text{ST}} (\dot{Q}_{\text{St, Gas}} + \dot{Q}_{\text{St, WHB}}) = \eta_{\text{ST}} (\beta \cdot \dot{Q}_F + \eta_{\text{WHB}} \cdot \eta_{\text{Gas}} \cdot (1 - \eta_{\text{GT}}) \cdot \dot{Q}_F) \quad (7.20)$$

The efficiency of the IGCC process as a whole is then given as

$$\eta_{\text{IGCC}} = \eta_{\text{Gas}} (\eta_{\text{GT}} + \eta_{\text{ST}} \eta_{\text{WHB}} (1 - \eta_{\text{GT}})) + \eta_{\text{ST}} \cdot \beta \quad (7.21)$$

Using Eq. (7.21), it is possible to discuss the influence of the gasification efficiency,  $\eta_{\text{Gas}}$ , on the total efficiency,  $\eta_{\text{IGCC}}$ . If the gasification efficiency  $\eta_{\text{Gas}}$  deteriorates, less output is produced in the gas and steam turbines and the efficiency of the IGCC process decreases. Usually an effort is made to utilise the losses occurring in the gasification island for steam production, the measure of this being the parameter  $\beta$ . This steam, however, is converted into electrical power only in the steam turbine, not in the more efficient gas and steam turbine process.

The efficiency of the gasification stage in an IGCC power plant means that compared to a natural gas fired combined cycle process, the overall efficiency is lower. State of the art in gas cleaning are processes at low temperatures, so that the gas tur-

bine is not supplied with sensible heat but only with chemical energy of the syngas. The gasification efficiency,  $\eta_{\text{Gas}}$ , corresponds to the cold gas efficiency,  $\eta_{\text{Cold Gas}}$ . Using a hot gas cleaning process it would be possible to increase the efficiency by providing the combined cycle process with additional sensible heat. An additional aim should be to convert the sensible heat into chemically bound energy. That is why the evaluation of the energy conversion in gasification processes generally brings into play cold gas efficiencies. These are typically at about 80%, with high-temperature gasifiers having lower and fluidised bed gasifiers having higher cold gas efficiencies.

The cold gas efficiency includes the carbon conversion, which is also used for drawing comparisons between gasifiers:

$$\eta_{\text{carbon}} = 1 - \frac{c_{\text{Res}}[\text{kmol/h}]}{c_{\text{Fuel}}[\text{kmol/h}]} \quad (7.22)$$

where  $c_{\text{Res}}$  is the carbon in the gasification residue in kmol/h and  $c_{\text{Fuel}}$  is the carbon in the fuel in kmol/h. The carbon conversion in high-temperature entrained-flow gasifiers reaches about 99%; in fluidised bed gasifiers, however, the conversion of carbon is lower and is a function of the reactivity and the volatile matter content of the fuel.

A further reduction of the efficiency of IGCC power plants comes about as a result of the energy used by the air separation process. By integrating the air separation unit into the process as a whole, the input energy can be reduced or energy can be recovered.

For natural gas fuelled IGCC processes, efficiencies of 50–60% are the current state of the art. This is the result of Eq. (7.21) if the efficiency  $\eta_{\text{Gas}}$  is set at 1 and the steam fraction  $\beta$  is set at 0. The gas turbine efficiency,  $\eta_{\text{GT}}$ , ranges around 40%; the efficiency of the steam process,  $\eta_{\text{ST}}$ , is about 35% and the efficiency of the heat recovery steam generator,  $\eta_{\text{WHB}}$ , about 90%. So the efficiency of the combined cycle,  $\eta_{\text{GuD}}$ , reaches 59% as a result. If the coal gasification efficiency is around 80%, the IGCC efficiency falls to 47% if no steam is produced. Assuming all losses are used for steam production ( $\beta = 0.2$ ), the efficiency could be increased by a maximum of 7%. In practice though, the extra efficiency is significantly lower, being partly used up by the air separation process. These considerations clarify the need to achieve a cold gas efficiency as high as possible.

### 7.6.3 Gasification Systems and Chemical Reactions

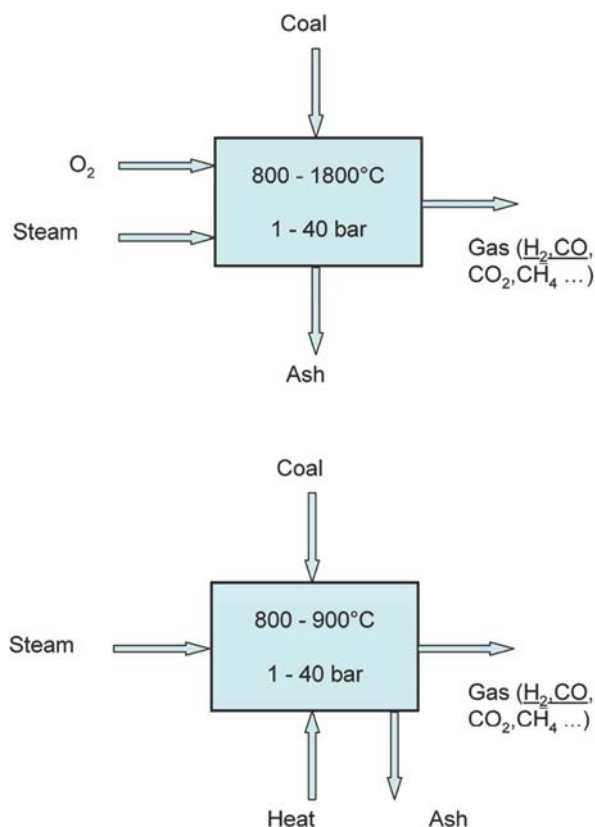
#### 7.6.3.1 Allothermal and Autothermal Gasification

Gasification systems can be categorised according to the heat input into autothermal and allothermal gasification processes (Jüntgen and van Heek 1981).

The principle of autothermal gasification using steam is depicted in a simplified form in Fig. 7.70. The fuel, depending on the reactor type, is converted at temperatures between 800 and 1,800°C by means of oxygen and water vapour into

a gas which, besides  $\text{CO}_2$  and perhaps also  $\text{CH}_4$ , mainly contains  $\text{CO}$  and  $\text{H}_2$ . The mixture of the latter two gases is termed synthesis gas because it is used in the chemical industry for the synthesis of methanol, ammonia and hydrocarbons (the latter using the Fischer – Tropsch process). The term autothermal comes from the fact that the process runs without external heat supply and because the heat consumption of the endothermic reaction of the coal with steam and the heat generation of the exothermic reaction with oxygen compensate each other. If air is used instead of oxygen, the gas produced becomes diluted by nitrogen and is usually only suited to be burned on the spot for heating purposes. One disadvantage of autothermal gasification is the lower gasification efficiency due to burning part of the coal. Furthermore, the production of a highly calorific gas needs oxygen as a feedstock and thus a more complex air separation plant.

The principle of allothermal gasification is also shown schematically in Fig. 7.70. Heat in this case is externally supplied to meet the heat requirement of the endothermic gasification reaction of the coal with water vapour. The heat can be fed to the gasification medium by recuperative tube-bundle heat exchangers or regeneratively via a circulating heat-carrying medium. The heat transfers from heating medium to tube wall and from tube wall to the fuel (recuperative heat exchanger) or



**Fig. 7.70** Principle of autothermal (*above*) and allothermal gasification (*below*)

to and from the heat-carrying medium (regenerative heat exchanger) are parameters determining the size and economy of the process. The fact that fluidised beds are characterised by a good heat transfer is the reason why they are taken into particular consideration for allothermal gasification. For greater gasification capacities, however, allothermal gasification is yet to be accepted because of the heat transfer problems. The method is practical for smaller capacities because the additional work and expense of heat transfer is compensated by not having an air separation step.

### 7.6.3.2 Basic Chemical Reactions

In gasification, similar to combustion (see Sect. 5.1), different phases are distinguished:

- Heating-up of the fuel
- Drying of the fuel
- Pyrolysis reactions
- Conversion of the solid carbon by gasification in the presence of oxygen, water vapour or  $\text{CO}_2$

In the same way as in combustion processes, heat has to be supplied for the partial processes of heating-up, drying and pyrolysis. The dominant factor of the gasification process is the heat demand of the gasification reactions. For this purpose, a corresponding amount of heat has to be fed from outside the process or released by partial oxidation.

In the entire process of coal gasification, numerous reactions take part. On the one hand, there are heterogeneous reactions in which the gasifying medium and also the product gases react with the solid matter. On the other hand, homogeneous reactions take place in the gaseous phase where the primary gaseous products go through conversion reactions involving both each other and the gasifying agent. The final composition of the gas produced depends on the interaction of all these reactions.

Despite the very complex molecular structure of coal, it is reasonable when discussing the reactions of the coal gasification to consider carbon alone as a first approximation, and only then, as a second approximation, the pyrolysis which precedes the gasification process. In the following, the most important heterogeneous and homogeneous reactions of carbon and of the pyrolysis are compiled. These can be used as basic reactions for describing the complex gasification reactions.

The principle possible pathways for gasifying carbonaceous fuel or fuel-containing hydrocarbon(s) are the partial oxidation reaction according to Eq. (7.23), the Boudouard reaction (Eq. 7.26), the heterogeneous water gas reaction (Eq. 7.27) and the methanation reaction (Eq. 7.28), all compiled in Table 7.15. The Boudouard reaction is of secondary importance in gas production from coal but it is significant for the blast furnace process. In this process, the carbon dioxide formed from the combustion reacts when flowing through the layers of coke above to form carbon monoxide, which for its part becomes oxygenated to form  $\text{CO}_2$  by reducing the iron

**Table 7.15** Gasification reactions (Higman and van der Burgt 2008), (Jüntgen and van Heek 1981)

<i>Combustion reactions</i>		
	$\Delta h$	
$C + \frac{1}{2} O_2 \leftrightarrow CO$	-111 MJ/kmol	(7.23)
$CO + \frac{1}{2} O_2 \leftrightarrow CO_2$	-283 MJ/kmol	(7.24)
$H_2 + \frac{1}{2} O_2 \leftrightarrow H_2O$	-242 MJ/kmol	(7.25)
<i>Heterogeneous gasification reactions</i>		
Boudouard reaction		
$C + CO_2 \leftrightarrow 2CO$	+172 MJ/kmol	(7.26)
Water gas reaction		
$C + H_2O \leftrightarrow CO + H_2$	+131 MJ/kmol	(7.27)
Methanation		
$C + 2 H_2 \leftrightarrow CH_4$	-75 MJ/kmol	(7.28)
<i>Homogeneous gasification reactions</i>		
Homogeneous water gas reaction		
$CO + H_2O \leftrightarrow CO_2 + H_2$	-41 MJ/kmol	(7.29)
Steam reforming		
$CH_4 + H_2O \leftrightarrow CO + 3H_2$	+206 MJ/kmol	(7.30)
<i>Pyrolysis reactions</i>		
$C_1H_xO_y \rightarrow (1 - y) C + y CO + x/2 H_2$	+17.4 kJ/mol <sup>1)</sup>	(7.31)
$C_1H_xO_y \rightarrow (1 - y - x/8) C + y CO + x/4 H_2 + x/8 CH_4$	+8.1 kJ/mol <sup>1)</sup>	(7.32)

<sup>1)</sup> For gas coal:  $x = 0.874$ ,  $y = 0.0794$

oxides. The methanation reaction is of great importance in hydrogasification, i.e. the gasification of coal by means of hydrogen. The objective of hydrogasification is to produce methane directly from coal as a substitute for natural gas. In allothermal steam gasification, the dominant process is the heterogeneous and strongly endothermic water gas reaction. In order to procure the reaction heat for 1 kg of carbon for steam gasification, about 0.29 kg of carbon has to be burned into  $CO_2$ . For most industrial gasifiers heated autothermally, the gasification process can be described by means of the partial oxidation and heterogeneous water gas reactions.

The principle reaction of the gas phase is the homogeneous water gas reaction (Eq. 7.29), where the carbon monoxide (formed previously) and water vapour are converted into hydrogen and carbon dioxide in an exothermic process. This reaction is used downstream in other industrial processes, not only in gasifiers, to convert CO completely or partially into hydrogen and to create a suitable feed gas for syntheses or other kinds of use. This step is also termed conversion or the CO shift reaction.

Another important reaction is the endothermic steam-reforming reaction (Eq. 7.30), which serves to convert methane into a synthesis gas. Running in the reverse direction, it is a strongly exothermic methanation reaction, which is used in downstream catalytic processes to produce methane.

The combustion reactions with oxygen (Eqs. 7.23, 7.24 and 7.25) mostly run to completion under gasification conditions, so they need not be considered for determining the equilibrium. The three heterogeneous reactions (Eqs. 7.26, 7.27 and 7.28) suffice for this purpose. Assuming a complete conversion of carbon as

in the case of entrained-flow gasifiers, it is possible to transform the reaction equations (7.26), (7.27) and (7.28) in order to obtain the two reaction equations (7.29) (subtraction of reaction 7.26 from reaction 7.27) and (7.30) (subtraction of reaction 7.28 from reaction 7.27). In entrained-flow gasifiers, the temperatures are commonly so high that in terms of thermodynamics, and in practice, hydrocarbons are no longer able to be found in noticeable concentrations.

So far the first reaction phase in the process of heating the coal up to the reaction temperature – the so-called pyrolysis reaction – has been ignored. In this phase, the coal is decomposed to coke and liquid and gaseous substances, which, depending on the reaction conditions, continue to react with the gasifying agent to form the gasification products. In general, pyrolysis can be described as follows:



If the pyrolysis products in the gasification reactor convert into the successor products of gasification, the following simple reaction scheme is sufficient:



or for high-temperature processes:



Table 7.15 also includes the pyrolysis reactions, showing the possible processes of conversion into carbon and the gasification products CO and H<sub>2</sub> (Eq. 7.41) or CO, H<sub>2</sub> and CH<sub>4</sub> (Eq. 7.42). It should be noted that the analyses in Eqs. 7.41 and 7.42, in the same way as the previous reactions, are made on a molar basis. For a bituminous coal with a weight composition of 85% C, 6% H<sub>2</sub> and 9% O<sub>2</sub>, for instance, the result is a molar elemental formula of C<sub>1</sub>H<sub>0.847</sub>O<sub>0.0794</sub> and a molar weight of 14.12 kg/kmol. The pyrolysis reactions are slightly endothermic.

Besides the components of carbon, hydrogen and oxygen, most fuels also contain smaller fractions of sulphur and nitrogen. In the gasification process, sulphur is mainly converted into hydrogen sulphide; COS, CS<sub>2</sub> and other sulphur-containing molecules form only in small quantities. Fuel nitrogen is converted into molecular nitrogen, NH<sub>3</sub> and HCN. These compounds have little influence on the principle gas composition of the synthesis gas. Further information is found in Sect. 7.6.5.1.

### 7.6.3.3 Considerations of the Thermodynamic Equilibrium

Analyses of thermodynamic equilibrium help to calculate the reaction progress achieved therein, i.e. the greatest possible stable reaction progress of the reactions involved. For the reversible reactions in Table 7.15, chemical equilibrium, where forward and reverse reactions run at the same rate, is a function of temperature and pressure. Time, however, is not considered as a factor in such analyses, so information on how fast these states develop is not generated.



Quantitative information on the conversion and the composition of products is obtained by applying the law of mass action for equilibrium reactions. Each chemical equilibrium reaction can be formulated according to the general form

$$\sum_{i=1}^N \nu_i A_i = 0 \quad (7.36)$$

where  $\nu_i$  is the molar ratio, or stoichiometric coefficient, and  $A_i$  is the substance involved in the reaction.

The state of the chemical equilibrium is described by the law of mass action, which expresses the correlation between the parameters depending on the concentration (measured, for instance, as the partial pressure or the volume fraction). Using the partial pressures  $p_{Ai}$  of the substances involved, the result for the equilibrium constant  $k_p$  is

$$k_p = \prod_{i=1}^N p_{Ai}^{\nu_i} \quad (7.37)$$

The law of mass action can also be applied to heterogeneous reactions. However, since the steam concentration is independent of the concentration of solids, and depends only on the temperature, this fixed value is included in the equilibrium constants, so the concentration of the solids need not be taken into account.

For the temperature-dependent equilibrium constant  $k_p$ , the gas constant  $R$ , the temperature and the reaction enthalpy  $\Delta H$ , the following relation applies:

$$d \frac{\ln k_p}{dT} = \frac{\Delta H}{RT^2} \quad (7.38)$$

This means that, for endothermic reactions ( $\Delta H > 0$ ),  $k_p$  rises with rising temperatures, i.e. a temperature increase pushes the reaction in the direction of the products. Inversely,  $k_p$ , for exothermic reactions ( $\Delta H < 0$ ), rises with falling temperatures, so in this case a decrease in temperature is favourable for the formation of the products.

As the composition of the product gases is of interest in gasification, it makes sense to represent the law of mass action as a function of the volume fractions  $x_i$ . The equilibrium constant  $k_x$  describes the correlation of the volume fractions  $x_i$ :

$$k_x = \prod_{i=1}^N x_i^{\nu_i} \quad (7.39)$$

Dalton's law gives the relation between the volume fraction  $x_i$  and the partial pressure  $p_{Ai}$ :

$$x_i = \frac{p_{Ai}}{p} \quad (7.40)$$

Consequently, the following relation between  $k_x$  and  $k_p$  applies:

$$k_x = \prod_{i=1}^N x_i^{v_i} = \frac{k_p}{p^{\sum v_i}} \quad (7.41)$$

This means that for reactions where the molar ratio  $\sum v_i$  is not changed, the equilibrium constants  $k_x$  and  $k_p$  are equal. An example of a consequence of this can be shown for the homogeneous water gas reaction with  $\sum v_i = 0$ :

$$k_p = \frac{p_{\text{CO}_2} \cdot p_{\text{H}_2}}{p_{\text{CO}} \cdot p_{\text{H}_2\text{O}}} = k_x = \frac{x_{\text{CO}_2} \cdot x_{\text{H}_2}}{x_{\text{CO}} \cdot x_{\text{H}_2\text{O}}} \quad (7.42)$$

In reactions with a changing molar ratio, the total pressure  $p$  has an additional influence on the equilibrium composition. For the heterogeneous water gas reaction with a change in the molar ratio  $\sum v_i$ , for instance, the expression is

$$k_x = \frac{x_{\text{CO}} \cdot x_{\text{H}_2}}{x_{\text{H}_2\text{O}}} = \frac{k_p}{p} = \frac{p_{\text{CO}} p_{\text{H}_2}}{p_{\text{H}_2\text{O}} p} \quad (7.43)$$

and for the steam-reforming reaction with a change in the molar ratio of 2, the expression becomes

$$k_x = \frac{x_{\text{CO}} \cdot x_{\text{H}_2}^3}{x_{\text{CH}_4} \cdot x_{\text{H}_2\text{O}}} = \frac{k_p}{p^2} = \frac{p_{\text{CO}} p_{\text{H}_2}^3}{p_{\text{CH}_4} p_{\text{H}_2\text{O}} p^2} = \frac{1}{p^2} \quad (7.44)$$

In both reactions, the equilibrium shifts towards the reactants with increasing pressure; with a temperature increase, it shifts in favour of the products because of the endothermic nature of the reactions. This corresponds to the principle of the minimal constraint developed by Le Chatelier, which says that a rise in pressure shifts the equilibrium in the direction of a volume decrease and a lowering of the pressure shifts the equilibrium towards a volume increase.

The equilibrium constants serve to calculate the conversion of the reactant material or the composition of the product gases. The equilibrium constants can be calculated either by experiment, using measured equilibrium concentrations, or using thermodynamic variables such as enthalpies and entropies.

Another, often applied, calculation method is the minimisation of the free enthalpy, also called the Gibbs free energy:

$$dG = dH - T dS \quad (7.45)$$

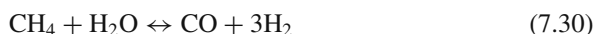
The method is based on the fact that in the state of equilibrium, a minimum of the free enthalpy arises. For the calculation of the free enthalpy of the mixtures, the thermodynamic data of the enthalpy and the entropy has to be known. Software and databases are commercially available for this purpose.

Thermodynamic analyses only describe the reality in gasification processes if the reactions run very quickly in comparison to the residence times. This does apply in some cases, for instance for the homogeneous reactions in entrained-flow reactors, due to the high temperatures of 1,500–1,600°C involved. Assuming an almost complete conversion of carbon, it is possible to calculate the gas-phase composition using Eqs. (7.39) and (7.40). In fluidised bed reactors, with low temperatures of 800–900°C, the gas composition cannot be determined by means of equilibrium analyses, though they may help to determine qualitative effects. The reactions involving the solid matter, in particular the gasification reactions of the solid carbon with steam, are the slowest reactions in the gasification process, so in this case kinetic analyses are required.

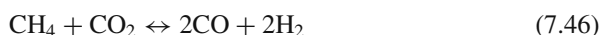
#### Effect of Pressure and Temperature on Gas Composition and Gasifier Efficiency

High pressures in gasification processes bear many advantages. This is why almost all industrial-scale gasifiers are operated at pressures of between 10 and 100 bar. High pressures reduce the energy demand of the total process, because the compressive work needed for the mass flows fed to the gasifier (oxygen, steam, fuel) is less than the work required for compressing the obtained synthesis gas. Furthermore, high pressures involve more compact and thus less expensive components. The pressure of the gasifier has to be adjusted to align with the rest of the process as a whole. For the exploitation of the syngas in combined cycle processes, pressures up to 30 bar are reasonable. For methanol synthesis, pressures from 130 to 180 bar would be advantageous, but such high pressures are not feasible in gasifiers.

Figure 7.71 shows the effect of the pressure on the gas composition from coal gasification at a temperature of 1,000°C based on thermodynamic equilibrium calculations. With rising pressure, the fractions of methane, carbon dioxide and steam increase; conversely, the contents of CO and H<sub>2</sub> diminish. This can be explained by the pressure effect, described above, on the steam-reforming reaction:



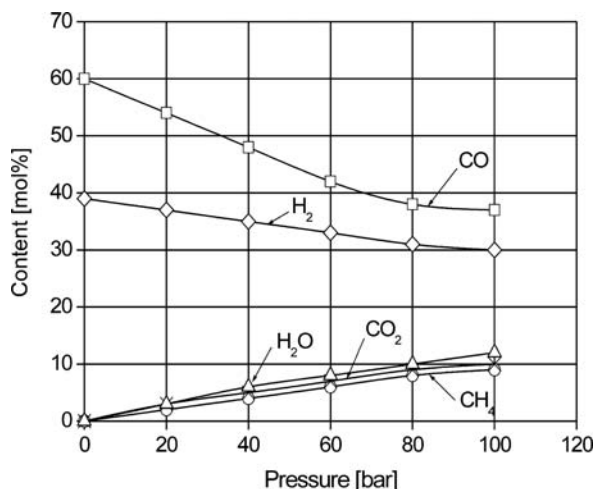
and on the reaction of methane with carbon dioxide:



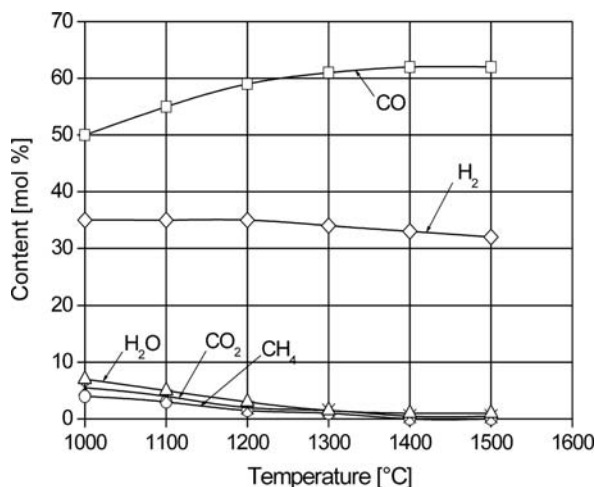
because the pressure works towards volume reduction. At higher temperatures, the effect of pressure is smaller, because in endothermic reactions higher temperatures shift the equilibrium in the direction of the products.

Figure 7.72 shows the effect of the temperature on the equilibrium composition at a pressure of 30 bar. As low methane contents are desired in the production of synthesis gas, temperatures higher than 1,300°C are required. For an IGCC process, these high temperatures are not necessary in this respect, because methane is rather advantageous. However, entrained-flow gasifiers are nevertheless designed for high

**Fig. 7.71** Variation of syngas compositions with pressure at a temperature of 1,000°C (from Higman and van der Burgt 2008, © 2008, with permission from Elsevier)



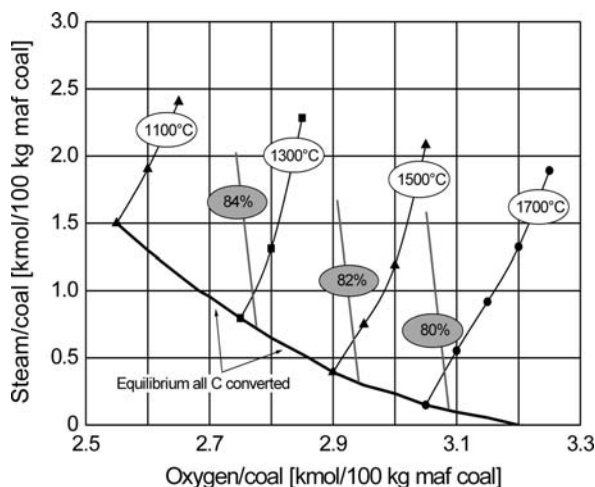
**Fig. 7.72** Variation of syngas compositions due to temperature at a pressure of 30 bar (from Higman and van der Burgt 2008, © 2008, with permission from Elsevier)



temperatures, because the fuel's ash fluid temperature has to be exceeded for safe operation with molten slag removal.

Knowing the composition of the generated gas product, the gasifier efficiencies can also be determined (assuming a complete conversion of the carbon). Figure 7.73 depicts the isolines of a complete carbon conversion for constant gasifier temperatures as a function of the necessary amounts of steam and oxygen. The higher the temperatures in the gasifier, the more the oxygen needed. Higher temperatures mean that the cold gas efficiency decreases, so in order to achieve an efficiency as high as possible for IGCC power plants, the temperature should be the lowest possible. For the entrained-flow gasifier, however, temperatures below 1,400–1,500°C, depending on the coal type, are impossible. The operating mode has to be adjusted so that, with

**Fig. 7.73** Cold gas efficiencies (from Higman and van der Burgt 2008, © 2008, with permission from Elsevier)



as little oxygen and steam as possible, the carbon is completely converted and the temperature can be reliably maintained during operation.

### 7.6.4 Classification of Coal Gasifiers

For coal gasification, a great number of methods have been developed which, in terms of process engineering, can be traced back to three basic principles. Analogous to combustion methods (see Chap. 5 and Fig. 5.1), the gas/solids reactor types employed can be categorised according to the state of motion of the solid matter:

- Fixed bed gasification
- Fluidised bed gasification
- Entrained-flow gasification (Strauß 2006; Higman and van der Burgt 2008; Jüntgen and van Heek 1981)

Table 7.16 gives an overview of the gasification methods and their characteristics.

#### 7.6.4.1 Fixed Bed Gasifiers

In a fixed bed reactor, the fuel is at rest. The gas flow can be conducted through the bed in co-, counter- or cross-flow. For better heat transfer, the flow of gas is usually conducted in a counterflow arrangement, as depicted in Fig. 7.74. In the direction of the fuel flow, three zones thus form in the fixed bed: a drying zone, a gasification zone and a combustion zone. In the direction of the gas flow, first the combustion of the solid carbon takes place; the rising hot gases then supply the gasification zone (which lies just above the combustion zone) with heat for the conversion of the

Table 7.16 Characteristics of different gasification processes

Category	Moving-Bed	Fluid-Bed	Entrained-Flow
Ash conditions	Dry ash screw	Dry ash screw	Slagging slurry
Feeding	Lurgi	Winkler, HTW, CFB	Pneumatic Shell, Siemens E-Gas
Typical process			
<b>Feed characteristics</b>			
Size	6–50 mm	6–10 mm	< 100 µm
Acceptability of fines	limited	good	unlimited
Acceptability of caking coal	yes (with stirrer)	possibly	yes
Preferred coal rank	any	low	any
<b>Operating characteristics</b>			
Reactor temperature	800–1200°C	900–1000°C	> 1400°C
Outlet gas temperature	425–650°C	900–1000°C	1250–1600°C
Residence time	10–30 min	1–10 min	< 1s
CO <sub>2</sub> vol. % (dry)	5	20	2
CO vol. % (dry)	56	40	66
H <sub>2</sub> vol. % (dry)	28	35	28
N <sub>2</sub> + Ar vol. % (dry)	3	2	4
CH <sub>4</sub> vol. % (dry)	7	3	0
C <sub>n</sub> H <sub>m</sub> <sup>2</sup> vol. % (dry)	1	0	0
Oxidant demand	low	moderate	high
Steam demand	high	moderate	low
Cold gas efficiency	very high	high	low (82%)
Carbon conversion	high	low	very high
			low (72%)
			very high

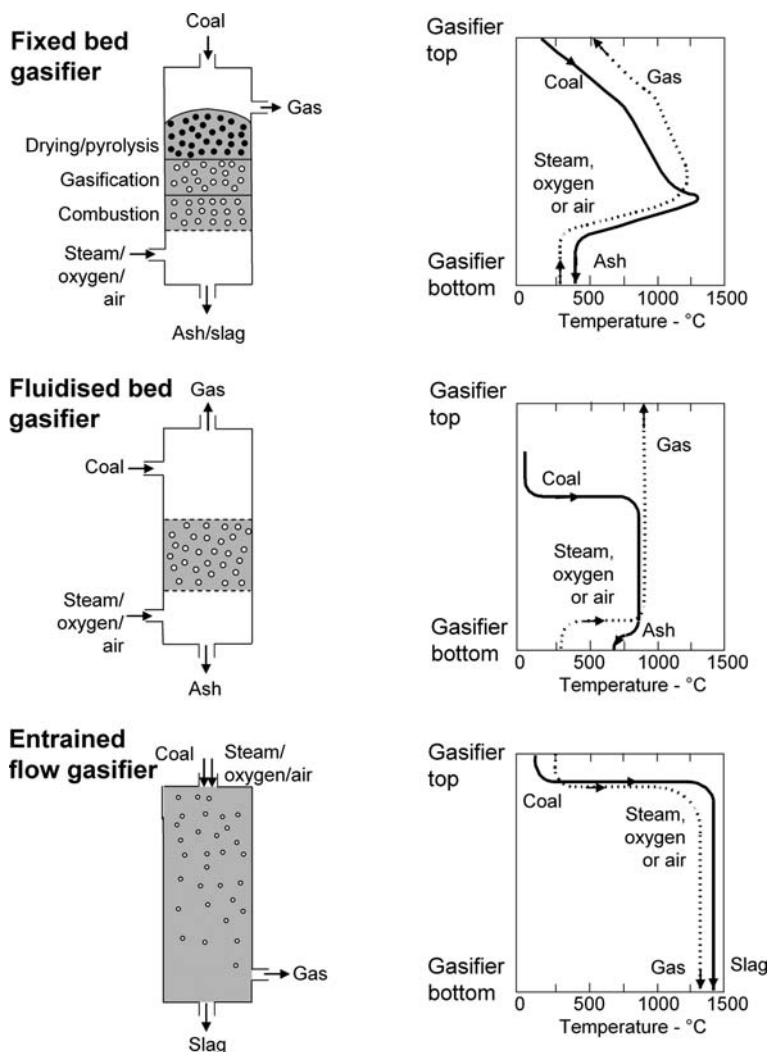


Fig. 7.74 Major types of gasifiers

solid carbon. Subsequently, the residual heat of the rising gases is used to pyrolyse the entering fuel and to dry it.

The advantage of the high level of heat recovery is countered by the disadvantage of the high tar content of the product gas – as the product gas cools down, the tar condenses. The fraction of the tar components can amount to 25% with respect to the supplied fuel energy. The gas produced exits the gasifier at a temperature of 400–500°C, with a content of  $\text{CH}_4$  as high as between 10 and 15%. If the energy content of the tar is used, the cold gas efficiency may be around 90%, but such waste

heat utilisation in a raw gas cooler is problematic because of the very fact of the high tar fractions.

Owing to the long residence time in the fixed bed, the carbon is almost completely converted, while consuming little oxygen. The coal types preferred are non-caking ones with a particle size ranging from 5 to 30 mm; the proportion of solid fuel fines has to be limited to ensure that there is a continuous flow through the fixed bed.

The method developed by Lurgi, called the Lurgi Dry Ash Process, goes back to a patent from 1927 and, for years, was the only pressurised gasification system. In Sasol, South Africa, 97 of a total of 152 fixed bed gasifiers worldwide are being operated at present, being used for the production of a synthesis gas for the Fischer – Tropsch process. A modified process is the British Gas/Lurgi slagging gasifier, which has been further developed for molten ash removal.

#### 7.6.4.2 Fluidised Bed Gasifiers

The operation of a fluidised bed is described in principle in Sects. 5.1 and 5.4, and illustrated in Fig. 7.74. The solid particles are either made to float by the upward-directed flow (stationary fluidised bed) or carried out and circulated by the flow (circulating fluidised bed). In the fluidised bed reactor, the intensive solids mixing means the temperature is nearly constant. The temperature in the fluidised bed is kept below the deformation temperature of the ash so that the ash does not stick together and can be removed in a dry state. In consequence, the fluidised bed temperature is specified as a function of the fuel and determines the carbon conversion. The typical temperatures for coals range between 950 and 1,100°C, while for biomass, they are between 800 and 950°C. Fine coal particles are carried out of the fluidised bed by the gas flow and thus diminish the carbon conversion. Typical carbon conversion rates lie between 95 and 97% and so are significantly lower than the rates in entrained-flow reactors. The advantages of fluidised beds compared to fixed bed gasifiers are the small contents of condensable by-products and the potential for use of coals with high ash contents. In contrast to the entrained-flow gasifier, milling is not needed, as it is sufficient to crush the coal to particle sizes smaller than 10 mm. The fluidised bed process is particularly suited to reactive coal types like lignite and for biomass; the cold gas efficiency ranges around 85%.

The atmospheric Winkler process was the first modern continuous gasification process with oxygen as the gasifying medium. The process was patented in 1922, and since then about 70 plants of that type have been constructed worldwide. Today, however, the process is only of historical interest, because almost all plants are no longer operated because of economic reasons. The gasifiers were operated at temperatures between 950 and 1,050°C and a fluidising velocity of 5 m/s. After a radiant cooler, the ash flow was removed along with a considerable amount of carbon, in the order of 20% of the fuel flow. Rheinbraun, in the 1970s, developed the high-temperature Winkler process (HTW), the essential characteristic of which was the upgrading of the fluidised bed process to handle pressures of up to 30 bar. A test plant of 600 t/day and a pressure of 10 bar was operated for more than 12



years with availabilities of 84%, the gas being used for methanol synthesis. Today, the HTW process is considered a possibility for IGCC power plants using lignite.

Circulating fluidised beds, with a more intensive gas – solids mixing and the inherent solids recirculation, have a number of advantages over stationary fluidised beds. There is, for instance, the higher carbon conversion rate and a performance insensitive to the particle size and form. The high gas velocities of 5–8 m/s ensure that most of the bigger particles are carried out and separated by the cyclone, then recirculated via a seal pot. Circulating fluidised beds have been developed by Foster Wheeler and Lurgi and are used today almost exclusively for biomass. In this process, air is used as the gasifying medium; the biomass has to be reduced to a size of 25–30 mm.

Allothermal fluidised bed gasifiers have the advantage of being able to produce a highly calorific product gas even without complex air separation. Developments of allothermal coal gasification processes, such as the *Mannesmann-Bergbauforschungsverfahren (MBG)* (“Mannesmann Mining Research Method”) in the 1970s and 1980s for example, have aimed at utilising high-temperature heat from high-temperature gas-cooled reactors (HTGR). In later concepts, heat produced from fossil fuels was incorporated into the fluidised bed by helium as a heat-carrying medium. With its necessarily large heat exchanger surfaces, however, the process failed to gain popularity. Two-stage allothermal processes, such as those sometimes used successfully for biomass, have not often been developed for or applied to coal gasification. The characteristic of the two-stage processes are separate reactors for combustion and gasification. In the combustion reactor, the residual coal from gasification is burned and the inert material of the fluidised bed is heated. The sensible heat of the sand, which serves as the heat-carrying medium, is used to gasify the fuel in a second fluidised bed. Two-stage gasifiers for biomass are described in Sect. 6.3.1.

### 7.6.4.3 Entrained-Flow Gasifier

In an entrained-flow process, finely milled coal with a particle size of less than 0.1 mm is fed directly into the reactor and converted in parallel flow with the gas at high temperatures and pressures, as shown in Fig. 7.74. The temperatures in the reactor are above the ash fluid point; depending on the calorific value of the coal type, they rise to between 1,400 and 1,600°C. The pressures of entrained-flow gasifiers in operation are between 20 and 70 bar. Given that the heat is provided through the combustion of part of the coal (i.e. an autothermal process), the cold gas efficiency lies below 85%. On account of the high temperatures, the inside walls of the gasifier are lined with a fireproof material; for high-ash fuels it is possible to do without such lining if the formation of a protecting slag layer on a cooled membrane wall can be ensured. The residence time of the coal particles in the reactor is about 1 s. In consequence, significantly higher volumetric power densities result for the entrained-flow reactor than for the other two reactor types. The carbon gets converted almost completely, the conversion rates amounting to more than 99%. Owing to the high reaction temperature, the entrained-flow process is also suitable

for less reactive fuels. However, expenditures of energy for the very fine milling have to be taken into account when considering the viability of this process for such fuels. High-moisture fuels, too, can be used, but the oxygen demand will rise and the cold gas efficiency will decrease in this case. About 60% of the ash is removed from the reactor in a liquid state, the rest being removed as fly ash from the product gas. Because of the high temperature, the product gas consists mainly of hydrogen and carbon monoxide (synthesis gas) and contains no tars.

### Process Variants

The advantages of the autothermal entrained-flow process – the high power density, the production of a tar-free synthesis gas and the fuel flexibility – have meant that the vast majority of gasifiers installed worldwide are autothermal entrained-flow types. The disadvantages of the high oxygen demand and lower cold gas efficiency can be limited through the selection of an appropriate process variant. The processes applied today are distinguished by the fuel feeding method (dry or wet), the cooling method for the reaction vessel (membrane wall or refractory lining) and the way the sensible heat of the hot product gas is used (a gas quench, water quench or a synthesis gas cooler). Furthermore, a distinction is made between one-stage and two-stage gasifiers. The design has a considerable influence on the cold gas efficiency and the overall efficiency, in which the effects can differ for IGCC and chemical applications. In the following, attention shall be given to IGCC applications with respect to the following:

- Wet or dry coal feeding: Wet coal feeding or using very moist fuels diminishes the cold gas efficiency, because water has to be vaporised and heated and because the oxygen demand increases. The more a fuel gets oxidised, the lower the cold gas efficiency will be. Wet coal feeding has the advantages of being simpler and having the ability to achieve very high pressures of up to 200 bar. Table 7.17 shows a comparison (Radtke et al. 2005; Uhde 2008).

Product gas cooling: The product gas heat can be utilised by means of a radiant cooler, which comes in the form of a water- or gas-quench cooler or a chemical quench. A radiant cooler produces steam which is used in the steam process of the combined cycle power plant. As this heat is not fed to the gas turbine, the conversion process runs at the lower efficiency of the Rankine process, so the efficiency is lower than for a natural gas fired combined cycle process. A gas quench in the following radiant cooler lowers the temperature of the waste heat utilisation process, for instance by quenching with recirculated product gas. As long as the remaining heat contained in the product gas is sufficient to achieve high steam temperatures, there are no negative effects from the radiant cooler. For the water quench, the same observations as for the gas quench hold true, but an additional drawback arises through the vaporisation of the water. The energy supplied for the vaporisation loses usefulness as it drops to the temperature of the condensation heat. Most satisfactory from the energetic point of view is a chemical quench, where the residual heat propels an endothermic reaction, and

**Table 7.17** Gas quality of dry and wet feeding (Radtke et al. 2005), (Uhde 2008)

	Dry Feed EF Gasifier	Slurry Type EF Gasifier
Coal/Petcoke feed	Dry pneumatic	Water slurry
Gasifier	Membrane wall	Refractory lining
Syngas cooling	Gas quench and convective cooler for IGCC applications	Water quench or radiant and convective cooler. (Radiant cooler is a very large piece of equipment.)
Solids removal	Almost completely dry	Wet scrubbing
Water treatment	Simple	Water quench is a potential source of corrosion, causes formation of formic acid, high water treatment costs.
Cold gas efficiency	82%	72%
Oxygen Consumption	0.93 kg/kg coal	1.07 kg/kg coal
Typical gas analysis vol. %		
CO <sub>2</sub>	2–3	15
CO	62	32
H <sub>2</sub>	27	27
H <sub>2</sub> S + COS	0.7	0.9
Inerts	5	1
H <sub>2</sub> O	2	23

the chemical energy of the product gases, or the cold gas efficiency, increases. This corresponds to concept designs that use two-stage gasifiers.

- Two-stage gasifiers: The purpose is to cool the product gas exiting the gasifier from temperatures between 1,500 and 1,600°C down to temperatures of about 1,000–1,100°C. While cooling the product gas, the heat extracted is used to propel the endothermic reactions of gasification, meaning that coal can be fed without oxygen to the second stage of the gasifier, bypassing the first. The lower temperatures mean that longer residence times for gasification are necessary and there is a risk that pyrolysis products do not react completely. For this reason there is a lower limit to the temperature. Carbon that has not reacted can be removed and recirculated after the product gas cooler. In another variant, incomplete gasification takes place in the first stage followed by an addition of steam for the complete gasification of the carbon in the second stage. Two-stage gasifiers have higher fractions of CH<sub>4</sub> and CO<sub>2</sub>. The decisive factor for the cold gas efficiency of two-stage gasifiers is the outlet temperature of the gasifier. A two-stage gasifier with an outlet temperature of 1,100°C has an efficiency corresponding to the cold gas efficiency of a one-stage gasifier with the same outlet temperature.
- Cooling of the gasifier: Gasifier cooling has the same effect as product gas cooling by steam. The heat removal is generally very low, though.

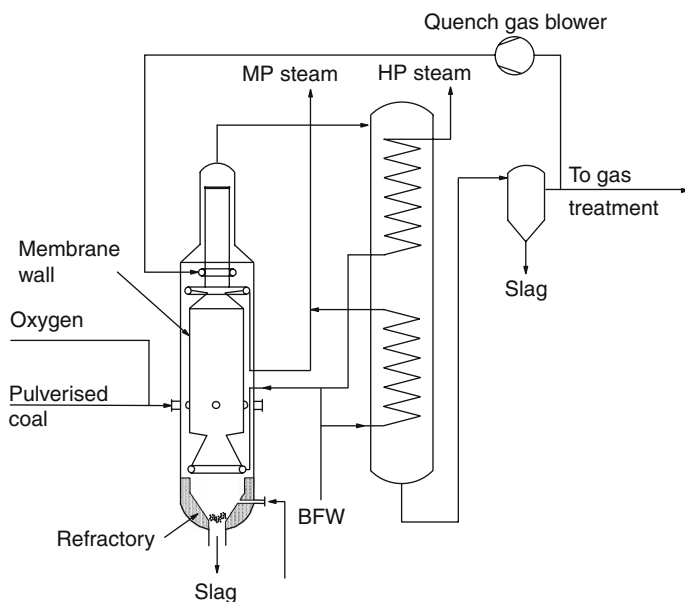
## Technologies

The development of entrained-flow gasification began with the development of the atmospherically operated Koppers-Totzek process in the 1950s. Commercial plants were operated in a great number of countries, mainly for ammonia synthesis. In recent years, no additional plants that follow this method have been built. Based on the Koppers-Totzek process, Shell and Koppers developed pressurised versions, first in co-operation and later separately. Both the Shell Coal Gasification Process (SCGP) and the Prenflo Process by Krupp-Koppers (later: Krupp-Uhde, today: Uhde) are one-stage entrained-flow gasifiers with dry coal feeding. Both designs feed coal in a dense flow to four burners using an inert gas. During the upward flow through the bed, where 90% of the particles are smaller than 90  $\mu\text{m}$ , the gasification of the fine coal particles takes place, with the molten ash flowing down into a water bath where it granulates. The temperatures in the gasifier are typically around 1,500°C and the pressure at about 30–40 bar. The reactor wall is a membrane wall construction that is studded and covered with a castable refractory mix in order to protect the metal wall from direct radiation and from the liquid slag. The tubes of the membrane wall are steam-cooled. The heat losses depend on the size of the reactor and on the quality and amount of slag and typically lie in the order of magnitude of 2–4% of the fuel heat. The hot product gas gets cooled down to 900°C by recirculated cold product gas at 280°C before exiting the gasifier. By means of this fast cooling down, cutting through the temperature range between the ash deformation and ash fluid temperatures, an attempt is made to prevent the formation of agglomerations. In the steam-cooled raw gas cooler, the product gas gets cooled from 900 to 280°C, afterwards being conducted to a particulate removal unit via ceramic filters. About half of the product gas is conducted to the quench cooler by use of a compressor, while the other half is led to a wet scrubber, becoming the net product. The Shell Coal Gasification Process is shown in Fig. 7.75.

In 1994, a 2,000 t/day Shell gasification unit was built for Demkolec (now NUON) in Buggenum in the Netherlands, using coal as feedstock. In 1997, Krupp-Koppers (now Uhde) built a 3,000 t/day unit for Elcogas in Puertollano, Spain, using a blend of high-ash coal and petcoke as the feedstock.

The Siemens gasification process is also a one-stage gasification method using a dry fuel supply. It can be traced back to developments of lignite gasification by the *Deutsches Brennstoffinstitut* in Freiberg in the 1970s. The entire fuel flow, steam and oxygen are fed through a burner. The reactor has a downward flow, with both molten slag and the hot product gas being discharged at the bottom (see Fig. 7.76). The discharge of slag and product gas together avoids a blocking of the slag out-flow. Depending on the application, partial quenching or full quenching may be employed. For high-ash fuels, a cooling screen lined with SiC is used, becoming covered by a protective slag layer during operation. For low-ash fuels, the design incorporates refractory lining.

The GE (formerly Texaco) process for coal gasification uses a slurry-feed downward-flowing entrained-flow gasifier. The coal is wet milled to a particle size of about 100  $\mu\text{m}$  and slurried in conventional equipment, then charged to the reactor with a membrane pump. The reactor pressure is typically about 30 bar for IGCC



**Fig. 7.75** The Shell Coal Gasification Process (from Higman and van der Burgt 2008, © 2008, with permission from Elsevier)

applications; for chemical applications it may be as much as 70–80 bar. The reactor shell is an uncooled refractory-lined vessel.

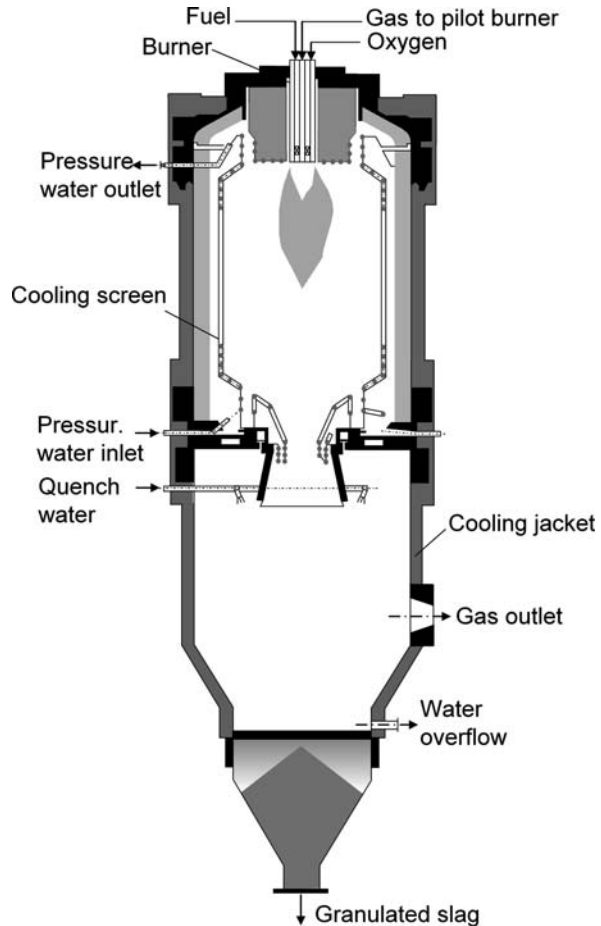
Syngas cooling can be performed by a radiant boiler or via a total water quench. In the quench configuration, the hot syngas leaves the reactor at the bottom together with the liquid ash and enters the quench chamber. The gas leaves the quench chamber fully saturated and at a temperature of between 200 and 300°C, which are suitable conditions for direct CO shift conversion. In the radiant cooler configuration, which is part of the Cool Water and Polk IGCC plants, full use is made of heat recovery for maximum efficiency. The GE gasifier is the most inexpensive gasifier on the market, but is maintenance-intensive. To achieve high availabilities of production, a standby reactor is required.

The E-Gas gasifier is currently the only two-stage process with an operating commercial-scale demonstration plant. A sub-bituminous coal – water slurry is injected into the hot gases coming from the first slagging stage, resulting in a temperature drop from 1,400 to 1,040°C. Unreacted char is separated by metallic candle filters and reinjected into the first slagging stage.

### 7.6.5 Gas Treatment

The composition of the raw gases produced by gasification differ considerably depending on the production method. For example, from gasification in a fluidised bed or an entrained-flow reactor, a raw gas has a high particulate content, whereas

**Fig. 7.76** Siemens gasifier with cooling screen (Source: Siemens Fuel Gasification)



from gasification in a fixed bed, the gas contains less particulates but a considerable amount of condensable pyrolysis products. Trace concentrations of pollutant gases depend on the fuel used and the gasification technology. The aim is to bring dry gas cleaning processes, using hot particulate removal, desulphurisation and dehalogenation, to a level considered state of the art. Such methods, however, have not yet been developed for application at an industrial scale. Wet gas cleaning, already tried and tested, is complex to implement and has a negative effect on the efficiency, because during the necessary cooling of the raw gas, about 15–20% of the supplied energy passes into the steam – water cycle (Maurstad 2005; Higman and van der Burgt 2008).

Besides depending on the gasification process and on the fuel, the necessary gas treatment steps depend on the purity requirements of the total process or on the syn-gas purity requirements. Correspondingly, the necessary gas treatment procedures differ, but in general comprise the following steps:

- Raw gas cooling of the syngas
- Gas cleaning of pollutant components such as particulates, sulphur, chlorine, nitrogen
- Gas preparation to adjust the composition of the product gas to that desired for downstream use

### 7.6.5.1 Impurities in the Gas

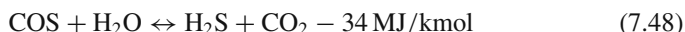
*Sulphur compounds:* Sulphur compounds in raw synthesis gas act as a catalyst poison for most chemical applications and for the low-temperature shift reaction. Used in IGCC, if untreated, these compounds are oxidised in the gas turbine and emitted as SO<sub>2</sub>.

In high-temperature gasification, over 90% of the sulphur components in the feedstock are converted to hydrogen sulphide (H<sub>2</sub>S) and the rest to carbonyl sulphide (COS). Compounds such as SO<sub>x</sub> and CS<sub>2</sub> are essentially absent in the syngas. This is not the case in low-temperature gasification, such as in the fixed bed process, where tars and other species are not completely cracked.

The relationship between H<sub>2</sub>S and COS contents of a raw gas is determined by the hydrogenation reaction:



and the hydrolysis reaction:



the rest being CO<sub>2</sub>.

Up to 99.8% of the sulphur can be removed in the acid gas removal process. As COS is not easily removed, a catalytic hydrolysis unit of COS to H<sub>2</sub>S prior to the acid gas removal is required.

*Nitrogen compounds:* Nitrogen enters the gasifier both as molecular nitrogen (supplied with the coal or oxygen flow) and as organic nitrogen in the fuel. The bulk of the nitrogen in the syngas is present as molecular nitrogen. Hydrogen cyanide (HCN) and ammonia (NH<sub>3</sub>) are produced in gasification in small amounts; due to the reducing conditions NO<sub>x</sub> is negligible. Most of the HCN and NH<sub>3</sub> originate from fuel-bound nitrogen, not from the molecular nitrogen, which has strong chemical bonds. The proportions of HCN and NH<sub>3</sub> are dependent on the coal characteristics. For the formation of NH<sub>3</sub> and HCN in a gasifier, the same fundamentals of NO<sub>x</sub> formation as discussed in Sect. 5.2.1 apply; however, due to the reducing conditions the oxidation is inhibited. Typical concentrations of HCN and NH<sub>3</sub> are in the low ppm range, up to several tens of ppm.

If untreated, both ammonia and hydrogen cyanide in the raw syngas would result in NO<sub>x</sub> emissions from IGCC processes. HCN can react with the amines used in the acid gas removal (AGR) unit and degrade them, while also being a poison for some catalytic processes (e.g. the Fischer – Tropsch synthesis). Both HCN and NH<sub>3</sub>

have very high solubilities in water and may therefore be easily removed by water scrubbing.

*Chlorine compounds:* Chlorine released from the coal is converted to hydrogen chloride (HCl) gas, which in contact with metals can form chlorides, such as sodium chloride (NaCl), with melting points in the range of 350–800°C. These metal chlorides pose a fouling risk in heat exchangers. Chlorine compounds from the coal will also react with ammonia to form ammonium chloride (NH<sub>4</sub>Cl) which is in the vapour phase at higher temperatures and becomes a solid at around 280°C. In addition to fouling problems, chlorine is a catalyst poison for the low-temperature shift reaction. Many of the chlorides may be removed in a water scrubber.

*Solid carbon and ash:* Ash and a small amount of remaining char will always be entrained in the exit flow of the gasifier. The quench or syngas cooler has to ensure that these particles will be non-sticky so that fouling problems are prevented. After capture in a filter or scrubber, char can be recycled to the gasifier to increase the carbon conversion efficiency.

*Other trace components:* Besides the major components, a variety of trace elements such as lead (Pb), mercury (Hg) and arsenic (As) are present in coals. Metal carbonyls such as nickel carbonyl (Ni(CO)<sub>4</sub>) and iron carbonyl (Fe(CO)<sub>5</sub>) can be formed.

### 7.6.5.2 Raw Gas Cooling

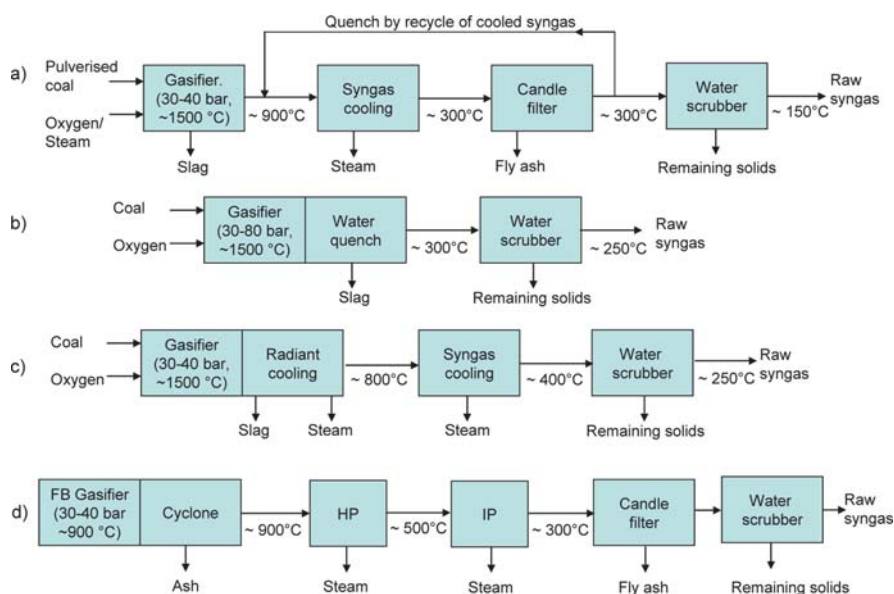
Raw gases exit gasifiers at high temperatures, ranging from 550°C from a fixed bed gasifier to 1,600°C from an entrained-flow gasifier. As the cleaning processes for the removal of these pollutants run at lower temperatures, the syngas needs to be cooled down. This cooling is required even for hot gas (or more correctly warm gas) cleaning. Figure 7.77 shows configurations for raw gas cooling and particulate removal units for different gasification systems.

Particular attention has to be paid to the cooling from gasification temperatures to temperatures below 900°C, because the ash is liquid in this temperature range and may cause build-ups of slag. For the sake of the continued availability of the process, the raw gases should therefore be cooled as fast as possible down to around 900°C, where the ash is present in solid form. For cooling in this range, the following process variants are used:

- Radiant cooling
- Water quenching
- Gas quenching
- Chemical quenching

In a radiant cooler, the heat of the syngas is transferred to the water – steam-cooled walls by radiation. Due to the high heat transfer rate, saturated steam is produced exclusively. The radiant cooler is an expensive component and susceptible to fouling and slagging.





**Fig. 7.77** Process flow diagram for different gasification processes (Maurstad 2005) and additions (a: EF + gas quench, b: EF + water quench, c: EF + radiant cooling, d: fluidised bed)

A water quench uses the sensible heat of the syngas to vaporise the injected water. With a partial quench, the syngas is cooled down to 900°C and the sensible heat (but not the latent heat) can be utilised for steam production. With a full quench, no high-pressure steam is generated and the syngas is saturated with steam. Water quenches, and the full quench in particular, are disadvantageous for IGCC processes without CO<sub>2</sub> separation, because the heat of evaporation cannot reasonably be used at the low temperature of condensation. For an IGCC power plant with CO<sub>2</sub> separation, the addition of water shifts the equilibrium of the water gas reactions in the direction of higher H<sub>2</sub>/CO contents.

One example of the use of a gas quench is in the Shell gasifier, where syngas that has already been cooled is mixed with the 1,500°C (hot) untreated synthesis gas, thus cooling to 900°C. This way, the heat is used within an unproblematic temperature range. Apart from the increased power consumption of the recirculation process, no other negative efficiency effects arise for an IGCC power plant because only sensible heat, not condensation heat, is produced.

Chemical quenching is advantageous because the gasifier exit temperature is lowered and thus the cold gas efficiency increased (see also Sect. "Process Variants").

For the process of further cooling of the flue gas by convective heat exchangers, the factors that should be considered are particulate removal and the behaviour of condensing components. The latter can be in the form of tars in biomass gasification, ammonium chloride in coal gasification or, simply, water. Typically, this cooling stage lowers the temperature from 900°C to about 300°C. At 900°C, the ash particles are no longer sticky; at 300°C, deposits of NH<sub>4</sub>Cl do not occur. A distinction is made

between water-tube and fire-tube boilers, both of which are in successful service. Fire-tube boilers conduct the synthesis gases inside the tubes, with the water flowing on the outside of them; in water-tube boilers, the high-pressure steam is produced inside the tubes. Typical steam pressures range from 100 to 150 bar for both types, but water-tube boilers can also be designed for even higher pressures. Fouling has to be taken into consideration for both types, so adequate cleaning facilities have to be included in the design. Superheating of the steam is possible. In order to limit corrosion, high-alloy materials are used and the material temperatures are restricted to values of 500–600°C.

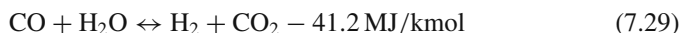
### 7.6.5.3 Particulate Removal

A dry particulate removal process should run at temperatures ranging between 300 and 500°C. Only at temperatures below 500°C do the alkali compounds achieve the maximum possible condensation on the fly ash, so that they are then removed along with the particulates. Below 300°C, the filters may be clogged by deposits of ammonium chloride. Cartridge filters are employed. By using topping cyclones prior to the filter, the filter load can be reduced.

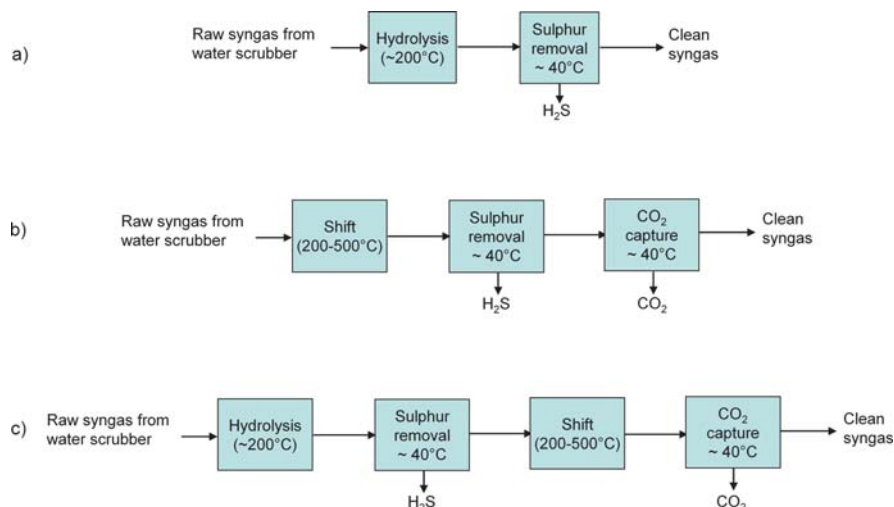
In most existing plants the (remaining) solids are washed out in venturi scrubbers or wash towers. The scrubbing takes place below the dew point of the gas, so that the finest solid particles can act as nuclei for condensation, thus ensuring that all solids are removed effectively. In wet scrubbing the water-soluble gaseous components such as  $\text{NH}_3$ ,  $\text{HCN}$ ,  $\text{HCl}$  and  $\text{HF}$  are also separated. The disadvantage of wet scrubbing is that the ash contains many regulated substances such as lead, zinc and cadmium, so the removed components must be disposed of carefully and according to relevant directives.

### 7.6.5.4 CO Shift

In an IGCC configuration with  $\text{CO}_2$  removal or in a hydrogen production plant, the water gas shift reaction is used to push the chemical composition of the syngas towards a maximum  $\text{H}_2$  yield:



According to the reaction, one mole of hydrogen can be produced from every mole of CO. The heating value per mole is less for  $\text{H}_2$  (241.8 MJ/kmol) than for CO (283.0 MJ/kmol), which means that chemical energy is converted to heat (exothermic reaction). The reaction itself is equimolar and therefore largely independent of pressure. The reaction is normally carried out in two stages, a high-temperature shift and a low-temperature shift. The high-temperature stage has the advantage of high reaction rates, while the low-temperature stage favours an equilibrium for maximum hydrogen production. Typical operating temperatures of the two stages are between 200 and 500°C, depending on the catalyst. The types of catalysts are distinguished by their operating temperature range and the maximum sulphur content



**Fig. 7.78** Process flow diagrams of gas cleaning (a) without shift conversion, (b) sour shift conversion, (c) clean shift conversion (Maurstad 2005)

of the syngas to be treated. The minimum molar  $\text{H}_2\text{O}/\text{CO}$  ratio is around 2. If there is not sufficient steam present in the syngas for the reaction, steam is extracted from the steam cycle.

Figure 7.78 shows the principle gas clean-up steps for processes with and without  $\text{CO}_2$  capture. If  $\text{CO}_2$  is not captured and the syngas is used to feed a turbine, then a shift is not required. In this case, a separate hydrolysis reactor is needed to convert COS to  $\text{H}_2\text{S}$  for easier sulphur removal. If there is a shift reaction, this conversion takes place simultaneously and no additional reactor is needed. For  $\text{CO}_2$  capture there are two alternative processes for the shift reaction:

- Sour shift (or raw shift)
- Clean shift

The sour shift is the preferred process when considering costs and efficiency. Gasifiers with a water quench are not suited to the clean shift, as a lot of valuable steam in the syngas would have to be condensed before sulphur removal and then, before the shift, a lot of steam would have to be added again. For a gasifier with dry gas quenching, the clean shift has some advantages, such as a cheaper catalyst and easier sulphur removal, as less  $\text{CO}_2$  is present. However, the more complex clean shift, with more heating and cooling, is less attractive from a capital cost and efficiency point of view for an IGCC plant (Maurstad 2005).

#### 7.6.5.5 Acid Gas Removal ( $\text{H}_2\text{S}$ , COS, $\text{CO}_2$ )

The term acid gas removal is often used as a synonym for desulphurisation, but strictly speaking, in the context of gasification, it also includes the acid gas  $\text{CO}_2$ . A

large number of different processes can be used for acid gas removal, which can be categorised according to the following principles used:

- Absorption or adsorption by a liquid solvent with a subsequent desorption step (chemical or physical washing)
- Absorption or adsorption on a solid material
- Diffusion through a permeable membrane

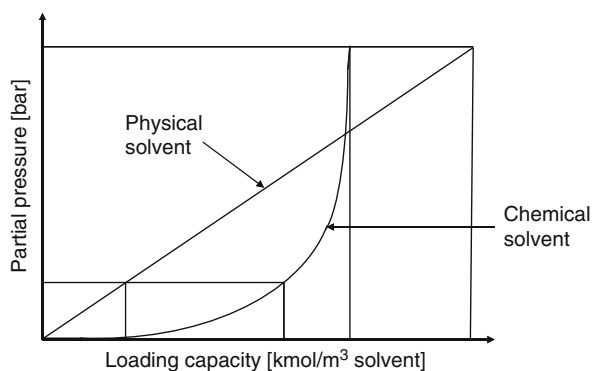
Acid gases such as  $\text{H}_2\text{S}$  and  $\text{CO}_2$  cannot be removed in a water wash process due to their low water solubilities. For acid gas removal, chemical (absorption) or physical (adsorption) washes with liquid solvents are normally used. The different principles for removal in gasification environments are illustrated in Fig. 7.79. The loading capacity of a physical solvent primarily depends on Henry's law and is therefore proportional to the partial pressure of the component to be removed. In contrast, the loading capacity of a chemical wash is limited by the quantity of the active component of the solvent. Generally, the solvent can be regenerated by flashing, stripping or reboiling or a combination of these. Both stripping and flashing reduce the partial pressure of the acid component and are used for physical solvents. In physical washes, reboiling raises the temperature and thus reduces the acid gas solubility. In chemical washes, the increased temperature breaks the chemical bonds and releases the components in the same chemical form in which they were absorbed.

Criteria for the selection of the appropriate process are as follows:

*Gas purity:* The demands of the syngas purity vary extremely with the application. For an IGCC power plant with a limit of 5 ppm  $\text{SO}_2$  in the flue gas, about 40 ppm  $\text{H}_2\text{S}$  at the outlet of the AGR is sufficient. For chemical applications such as ammonia, methanol or SNG production, 100 ppb may be required.

*Raw gas composition:* The washing solution must cope with the impurities in the raw gas. HCN in the raw gas, for instance, can react with amines, causing solution degradation.

*Selectivity:* The selectivity of a gas treatment process is the ability to remove  $\text{H}_2\text{S}$  while leaving  $\text{CO}_2$  in the synthesis gas.



**Fig. 7.79** Loading capacity of physical and chemical solvents (from Higman and van der Burgt 2008, © 2008, with permission from Elsevier)

Solutions of amines in water are commonly used for chemical washes; examples of amines are mono- and diethanolamine (MEA and DEA) and methyldiethanolamine (MDEA). MDEA is the most widely used amine today. Examples of physical washes are the Rectisol, Selexol and Purisol processes. The Rectisol process, which uses methanol as a solvent, operates between temperatures of  $-30$  and  $-60^{\circ}\text{C}$ . The process can achieve very high gas purities and is used for chemical applications where synthesis catalysts require sulphur removal to less than  $0.1$  ppmv. The Selexol process, which uses dimethyl ethers of polyethylene glycol (DMPEG), is operated at  $0$ – $40^{\circ}\text{C}$ , reducing refrigeration requirements, and can achieve gas purities of  $1$  ppm  $\text{H}_2\text{S}$  and  $\text{COS}$ , respectively. The Purisol process shows similar characteristics, but has a higher  $\text{H}_2\text{S}/\text{CO}_2$  selectivity.

Physical – chemical washes make use of the principles of both physical and chemical removal. They generally use an amine together with an organic physical solvent. An example of this is Shell's Sulfinol solvent; the modified m-Sulfinol solvent uses MDEA as the chemical component and is applied at the Buggenum plant.

### Sulphur Removal

The sulphur removal process consists of three process steps:

- Acid gas removal (AGR)
- Sulphur recovery (SR)
- Tail-gas treating (TGT)

The AGR process removes the  $\text{H}_2\text{S}$  from the syngas. In present-day IGCC plants, the two preferred processes are chemical washing, based on aqueous methyldiethanolamine (MDEA), and the Selexol process, based on a physical solvent. Both methods can reduce the total sulphur ( $\text{H}_2\text{S} + \text{COS}$ ) to levels below  $20$  ppmv in the cleaned syngas. For deep sulphur removal, required for chemical applications, the more expensive Rectisol process, using a physical solvent, may be applied. For  $\text{CO}_2$  capture a second-stage AGR has to be added to remove the  $\text{CO}_2$  from the sulphur-free syngas.

The purpose of the sulphur recovery unit (SRU) is to convert the  $\text{H}_2\text{S}$  into a chemical product which can be reused. The most common method for SRU is the Claus process which produces elemental sulphur by sub-stoichiometric combustion with air or oxygen. Different versions of this process are available. The sulphur may be fixed as elemental sulphur in liquid or solid form or as sulphuric acid. In order to achieve high enough degrees of sulphur recovery, the thermodynamics of the Claus process requires some treating of the tail gas, which usually contains mostly  $\text{H}_2\text{S}$  and  $\text{SO}_2$ , but also small amounts of  $\text{COS}$ ,  $\text{CS}_2$  and elemental sulphur vapours. In the TGT process, the sulphur species are converted to  $\text{H}_2\text{S}$ , which can then be absorbed in a liquid solvent.

## CO<sub>2</sub> Removal

CO<sub>2</sub> capture requires a second stage to the AGR process for the treatment of the sulphur-free syngas. A two-stage Selexol process is the preferred option for selective removal of sulphur and CO<sub>2</sub>. If combined capture of H<sub>2</sub>S and CO<sub>2</sub> is acceptable for a downstream storage or an enhanced oil recovery (EOR) project, significant cost reductions are possible because of a simpler AGR process and elimination of the SRU and TGT units in the sulphur removal process.

### 7.6.5.6 Hot Gas Cleaning

Hot gas cleaning units (HGCUs) remove particulates, sulphur compounds and other pollutants at higher temperatures than traditional processes such as water scrubbers and acid gas removal systems. HGCUs provide several advantages in comparison to state-of-the-art cold gas cleaning units (CGCU) operating at ambient temperatures or below:

- The total process efficiency increases, because syngas cooling is not required and water does not have to be removed from the syngas.
- Sour water treatment requirements are eliminated. Sour water is produced in CGCUs when syngas is cooled below the dew point of the water.
- Troublesome ash-char water mixtures produced in water quenching or wet scrubbing of particulates from the syngas can be avoided.
- There is the potential to reduce capital and operating costs (Korens et al. 2002; Holt 2003).

The development of hot gas clean-up systems has been pursued in the USA, Europe and Japan since the 1970s. The development has focussed primarily on syngas from air-blown gasification, because air-blown gasification systems produce over twice the volume of oxygen-blown systems due to dilution by nitrogen. Conventional cold gas cleaning for air-blown gasification incurs a costly and substantial efficiency loss, making it uneconomic. Therefore the success of air-blown gasification depends on the development of HGCUs. However, hot gas cleaning is also applicable to oxygen-blown gasification and improves its process efficiency in comparison to cold gas cleaning.

The temperature at which the product gas is used determines the temperature of the gas cleaning train. As most HGPU development programs have focussed on hot gas cleaning for IGCC applications, the temperature has been the highest possible at which the gas turbine fuel control and delivery systems can be designed. The requirement for very low alkali contents in the flue gas to prevent alkali corrosion of hot gas turbine components, and the desire to avoid expensive materials and unreliable refractory-lined pipes, sets this level at about 500–550°C. At this temperature the alkali vapour condenses on particles in the hot syngas which are then removed in the barrier filters. Since large-scale gasifiers operate at 1,400–1,600°C, significant cooling, the extent of which depends on the gasifier design and the feedstock, is still

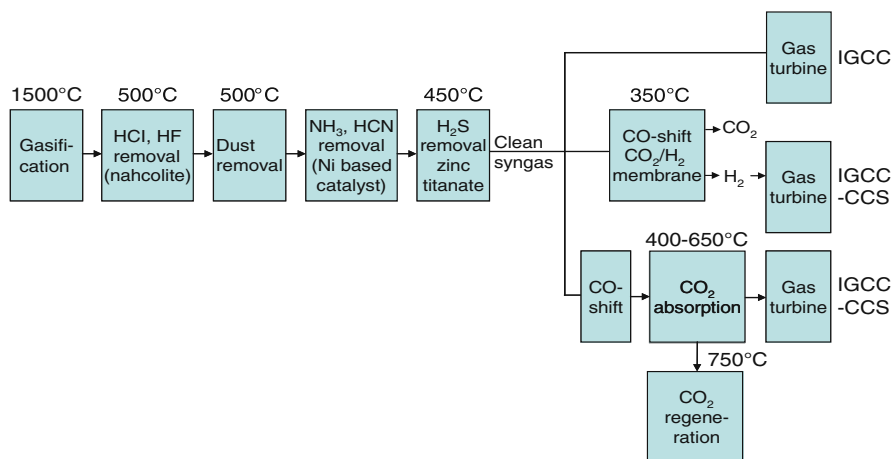


Fig. 7.80 Schematic diagram of a hot gas cleaning process

required. Most of the hot gas cleaning demonstration units have operated between 400 and 500°C, so that the term hot gas cleaning is misleading. Instead the term warm gas cleaning describes the actual temperature more appropriately.

Development of hot gas cleaning has focussed mainly on particle separation and removal of chloride, alkalis and sulphur components from syngas for gas turbine applications. In order to fulfil emission requirements, additional components such as HCl, HCN, NH<sub>3</sub> and mercury also have to be considered.

Figure 7.80 shows a schematic diagram of a possible hot gas cleaning process for an IGCC process with and without CO<sub>2</sub> removal. In the following, the principle process steps of hot gas cleaning are discussed and the state of development is described.

### Hot Gas Filtration

Barrier filters are the only currently commercially available HGC technology and have been successfully demonstrated in gasification projects. Candle filters – ceramic or metal tubes mounted in bundles, themselves within a filter vessel – are being used for final particulate removal for large syngas flows (refer to Fig. 7.18). The syngas flows from the outside through the porous tube walls, into the ceramic or metal tubes, and flows out of the vessel through the inside of the tubes. Back-pulsing the filtered gas dislodges the deposited ash from the outside of the candles (the ceramic or metal tubes), and the ash is discharged from the bottom of the vessel. The details are described in Sect. 7.3 in the context of pressurised fluidised bed combustion.

As described previously, the operating temperature of barrier filters is chosen to be below 550°C to promote the condensation of alkalis on particulates. The actual operation temperatures of the hot gas filters in the IGCC demonstration plants are

even lower. The temperatures of the hot gas filters are  $\sim 350^{\circ}\text{C}$  at Wabash (metallic elements) and  $250^{\circ}\text{C}$  at Buggenum (ceramic elements) and Puertollano. Solids bridging, candle degradation and breakage and fouling and corrosion of metallic components have been the major problems. Further improvements are still needed to increase the filter element lifetime and to reduce filter installation, operating and maintenance costs. Upstream cyclones are important components of a hot particle removal system – minimising the load on the hot filters. Overall, hot gas filtration offers definite advantages to IGCC over water scrubbing (Korens et al. 2002; Holt 2003).

### Alkali Cleaning

There are two main methods employed for cleaning vapour-phase alkali compounds:

- Cleaning at low temperatures. When the gas temperature is lowered below  $550\text{--}600^{\circ}\text{C}$ , alkali vapours condense and can be removed by particle removal systems.
- Syngases with alkali compounds can also be cleaned by passing through alkali getters, such as activated bauxite or activated alumina, at higher temperatures. Alkali compounds are then physisorbed or chemisorbed on the getter surface, with chemisorption suggested to be the dominant pathway when moisture is present. More details can be found in Sect. 7.4.3.

### Hot Gas Desulphurisation

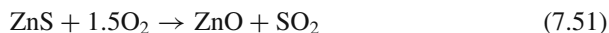
Metal oxide sorbents, which come as regenerable or disposable types, are able to capture  $\text{H}_2\text{S}$  at elevated temperatures. Disposable sorbents, such as limestone or dolomite, are typically calcium based and injected into the gasifier for in situ desulphurisation.  $\text{H}_2\text{S}$  reacts with these materials at  $950\text{--}1,050^{\circ}\text{C}$  and pressures over 20 bar to form  $\text{CaS}$ . Due to thermodynamic limitations, only 90% sulphur removal can be achieved, which means a typical outlet  $\text{H}_2\text{S}$  concentration of 300–500 ppmv. These sorbents can only be used once, which increases the amount of solid waste to be continuously removed from the process. Because  $\text{CaS}$  is not environmentally stable, it has to be converted to  $\text{CaSO}_4$ , which requires a separate oxidation stage (Atimay 2001).

Regenerable sorbents are usually used in a separate fixed or movable bed reactor after the gasifier. Having the unit separate makes it easier to regenerate the sorbent. Zinc oxide sorbents give the best results for  $\text{H}_2\text{S}$  cleaning at elevated temperatures (in the range  $350\text{--}750^{\circ}\text{C}$ ). The desulphurisation reaction is



and the regeneration reaction:





A great disadvantage of ZnO is the fact that it quickly reduces in reducing fuel gas atmospheres at high temperatures, so that the syngas is polluted with vaporised zinc. Zinc titanate can achieve the same residual H<sub>2</sub>S level, but is more stable and shows a better attrition resistance. In general, zinc titanate (Zn<sub>2</sub>TiO<sub>4</sub>) is considered the best option for H<sub>2</sub>S removal at high temperatures of up to 850°C (Aravind 2007).

The only two large-scale hot gas desulphurisation systems have been installed in the USA. They have never been demonstrated, however. Both systems were based on the reaction of H<sub>2</sub>S with zinc oxide/nickel oxide solid sorbents followed by regeneration of the sorbent by contact with air. The regenerator off-gas contained SO<sub>2</sub>, which had to be converted to elemental sulphur or sulphuric acid in a final recovery operation. At the 260 MW coal-fired IGCC in Tampa, the HGCU system was designed to treat 10% of the syngas flow. The HGCU was a moving bed absorption process designed for temperatures of 480°C. One of the reasons for cancelling the demonstration was the sorbent attrition behaviour, which led to extremely high annual sorbent costs. Interest in HGCU processes such as hot desulphurisation has been decreasing of late, partly because of disappointing results in finding solid sorbents with the necessary attrition resistance (Korens et al. 2002; Tampa Electric 1996).

### HCl Removal

Hydrogen chloride has to be removed from the syngas to prevent corrosion in the gas cleaning train and in the gas turbine. Sodium and potassium compounds are effective for dry removal of HCl and HF from syngas down to the ppm level at temperatures of 600°C. Higher temperatures result in an increase of gaseous alkalis (Aravind 2007). Nahcolite (naturally occurring sodium bicarbonate, NaHCO<sub>3</sub>) and sodium carbonate/bicarbonate mixtures are considered for use as sorbents. In the case of sodium carbonate, the following absorption reaction takes place:



If the sorbent is injected before the particle filter, the solid salt can be separated together with the dust.

### Mercury Removal

The prospect of stringent mercury emissions standards for coal conversion plants seriously dampens the outlook for hot or warm gas clean-up. It is believed that mercury removal becomes more difficult as the syngas temperature increases. If it is necessary to cool the syngas for mercury removal, then the motivation for hot

or warm gas desulphurisation disappears unless related economic benefits can be demonstrated (Korens et al. 2002).

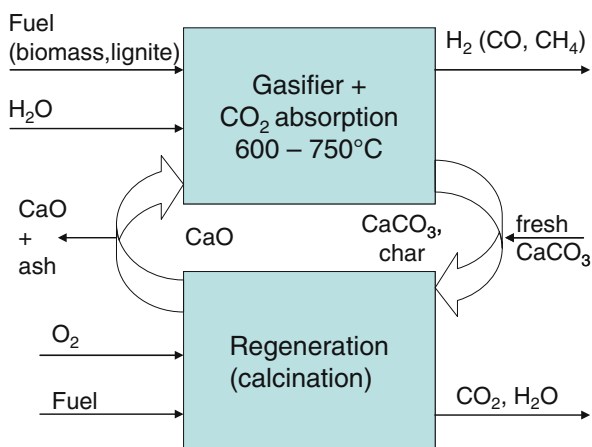
### Hot Gas Cleaning at Temperatures Above 1,400°C

For the process of pressurised pulverised coal combustion, comprehensive investigations have been carried out to develop a gas cleaning system at a temperature above the fluid temperature (1,400–1,600°C). Results of these developments have been described in Sect. 7.4. It was proven that it is possible to remove liquid slag and gaseous alkalis down to very low concentrations in the gas. It is assumed that it will also be possible to apply this technology to gasification systems, to remove liquid ash, alkalis and possibly heavy metals. If such cleaning is needed, the removal of sulphur and chlorine would require temperatures of about 600°C as described above. The higher temperature gas cleaning process would be beneficial for gasification systems combined with a membrane shift reactor. The requirements for such a reactor are not yet known. Investigations are currently being carried out in the framework of a research project (Müller et al. 2009; Spliethoff et al. 2009).

#### 7.6.5.7 CO<sub>2</sub> Separation at High Temperatures

Figure 7.80 shows a process configuration with hot gas cleaning and a CO<sub>2</sub> separation step at the end of the gas cleaning train, similar to corresponding arrangements for cold gas cleaning. For the removal of CO<sub>2</sub>, a water gas CO shift reactor is required prior to the separation to produce a H<sub>2</sub>/CO<sub>2</sub> mixture. There are two options for H<sub>2</sub>/CO<sub>2</sub> separation:

- CO/O<sub>2</sub> separation by means of high-temperature membranes. The membranes separate the syngas, already shifted to H<sub>2</sub> and CO<sub>2</sub>, into nearly pure streams of the two gases. In this process, however, the H<sub>2</sub> is produced at near-atmospheric



**Fig. 7.81** Sorption-enhanced reforming

pressure, requiring compression for use in IGCC or refinery processes. Most probably, any hot gas membrane would require the prior removal of all particulate material and other trace components that may be corrosive or plug the membrane pores, though the gas cleaning requirements depend on the membrane material and are not yet known. It has to be pointed out that this technology is far from mature and that further development is required before it is applied, mainly due to the high cost of ceramic membranes. Conceptual designs try to combine the functions of the CO shift and membrane separation processes in a water gas shift membrane reactor (WGSMR).

- Absorption of CO<sub>2</sub> by using solid sorbents at temperatures between 400 and 650°C and regeneration at 750°C. This process can be applied as a post-combustion capture technology (termed carbonate looping) and will be discussed in Sect. 8.5.2.

CO<sub>2</sub> absorption by CaO and regeneration of CaCO<sub>3</sub> can be combined with steam gasification (Weimer et al. 2008; Florin and Harris 2008). This process is known as lime-enhanced gasification of solid fuels (LEGS), sorption-enhanced reforming (SER) or absorption-enhanced reforming (AER). Figure 7.81 shows a schematic diagram of the process, which consists of a steam gasification reactor as well as a regeneration reactor. For both reactors, fluidised beds are proposed because of their excellent gas/particle contact and the fact that the convenient temperatures are appropriate for fluidised beds.

- In the gasifier, CaO absorbs CO<sub>2</sub> and increases the H<sub>2</sub> concentration of the product gas. In the case of biomass, H<sub>2</sub> concentrations of 40–50%, which are reported for steam gasification without CO<sub>2</sub> capture, increase to about 80% after CO<sub>2</sub> absorption. The process conditions of the gasification reactor have to be selected according to the carbon conversion and CO<sub>2</sub> absorption requirements. Carbon conversion is favoured at high temperatures, whereas the exothermic CO<sub>2</sub> absorption is favoured at lower temperatures. To achieve a carbon capture of 80% it is necessary to increase the pressure as the temperature increases, for example from a gasification pressure of 0.1 MPa at 650°C to 0.9 MPa at 750°C. Due to this reason atmospheric gasification and capture is only suitable for biomass, whereas the utilisation of brown coal requires pressurised conditions. The potential to further increase the pressure is limited, because increasing the pressure promotes methane formation and reduces carbon capture. The exothermic CO<sub>2</sub> absorption reaction provides heat for the endothermic gasification.
- In a second reactor, CO<sub>2</sub> is released by calcination of CaCO<sub>3</sub>, producing a high-purity CO<sub>2</sub> stream for storage. The regenerated CaO is recycled to the gasifier. At atmospheric pressure, the temperature required for regeneration is about 900°C. Higher pressures require higher temperatures for regeneration, which results in an energy penalty. The heat required for regeneration can be provided by the combustion of residual char from the gasifier. If a concentrated CO<sub>2</sub> stream is required, oxygen is needed for combustion.

## 7.6.6 Components and Integration

### 7.6.6.1 Gas Turbines

Gas turbines are designed for natural gas and liquid fuels, but are also commercially available for operation using syngas. The two coal IGCC demonstration plants in the USA (Tampa and Wabash) each use a GE 7FA turbine, while the two European plants at Buggenum and Puertollano use the Siemens SGT5-2000E (previously called V94.2) and the SGT5-4000F (previously called V94.3) (Maurstad 2005).

#### Syngas Combustion

Synthesis gas from high-temperature gasification essentially consists of carbon monoxide and hydrogen. Hydrogen is very reactive and increases the flame velocity, as opposed to fuels containing hydrocarbons, which decrease it. The calorific value of syngas from oxygen-blown gasifiers lies between about 8 and 11 MJ/kg, while it lies between 4 and 6 MJ/kg for air-blown gasifiers, which dilute the gas with nitrogen. These latter values are roughly 4–10 times lower than the calorific values of common gas turbine fuels. Given the lower air demand of  $H_2$  and CO, the adiabatic combustion temperatures – under standard conditions for the combustion air (1 bar, 15°C) – are around 2,040°C and thus 100°C higher than in the combustion of natural gas. By admixing the nitrogen fraction removed in air separation, the adiabatic combustion temperatures can be lowered to about 1,580°C. These flame temperatures are sufficiently low to limit the formation of thermal nitrogen in diffusion flames (Schetter et al. 1991).

Gas turbines which run on natural gas normally use the so-called dry low- $NO_x$  (DLN) combustors. In these combustion chambers, the fuel is pre-mixed with the total air to lower the flame temperatures and reduce the formation of thermal  $NO_x$ . They are designated “dry” because no injection of water or steam is used to reduce flame temperatures. However, DLN combustors are currently not used with syngas or hydrogen mixtures as fuels because of the danger of flashback caused by the high flame propagation speed of hydrogen. The current practice is to use traditional diffusion combustors, which require a diluent to reduce flame temperatures for  $NO_x$  control. The existing IGCC demonstration plants use nitrogen from the air separation unit, saturation of syngas with water or a combination of both to dilute the fuel before combustion with air. By these methods it is possible to reach a  $NO_x$  concentration of around 10 ppmv (at 15%  $O_2$ ) in the exhaust gas.

For coal gas burners, emissions have to be low both for coal gas and for natural gas firing. Figure 7.82 shows a standard hybrid burner and a modified hybrid burner, the latter of which is used in Buggenum and in Puertollano and operated as a simple diffusion burner. The syngas injection is via an annular swirl nozzle with a large cross-section around the centrally mounted natural gas or crude oil nozzle. The syngas flames in Buggenum are stable up to at least a calorific value of 4 MJ/kg and a 40% output and potentially more (Huth et al. 1998; Hannemann et al. 2003).

When a gas turbine is run on syngas, which has a higher share of hydrogen compared to natural gas, the exhaust gas may consist of more than the usual 8% by

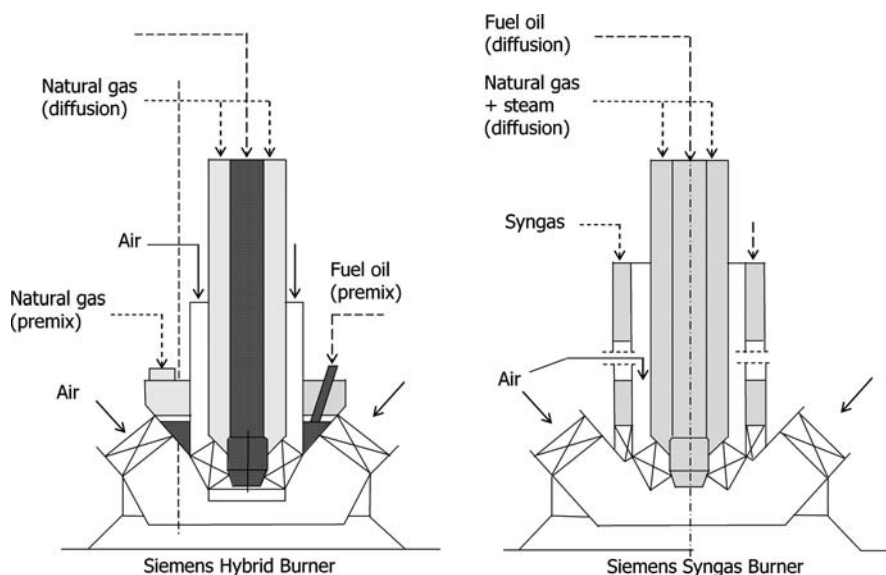


Fig. 7.82 A burner for syngas applications (Huth et al. 1998)

volume of water vapour. The water vapour concentration can vary between 5% (for the Puertollano fuel gas) and 14% (for pure hydrogen). The significance of a higher volume percentage of steam in the exhaust is an increased heat transfer, which raises the metal temperatures, thus shortening the lifetime of the turbine materials. Additionally, the increased mass flow through the turbine also results in an enhanced heat transfer.

While there are several gas turbines running on syngas, there are no turbines running solely on hydrogen. For pure hydrogen to be the fuel, the turbine inlet temperature (TIT) of the gas turbine would probably have to be reduced to avoid shorter lifetimes of the turbine blades (both the base materials and the coatings). This reduction in the TIT would reduce the efficiency of the combined cycle. In IGCC applications with  $\text{CO}_2$  removal, hydrogen can be diluted by water saturation or by nitrogen.

DLN burners for syngas or hydrogen are not state of the art, but their use would be beneficial, because they do not require such a massive dilution for  $\text{NO}_x$  control. Dilution with steam and nitrogen down to hydrogen concentrations of 50–70% is the countermeasure to control the high flame propagation speed. DLN burners are currently under development.

### Gas Turbine Power and Compressor Surge

As explained above, syngas use features a higher mass flow in comparison to natural gas. The increased mass flow of fuel and therefore the higher mass flowrate through the turbine will increase the power output from the turbine. If the fuel is diluted with nitrogen or water, the potential for increased GT power output is even higher. Depending on the turbine, there may be several limitations to the realisation of the

increased power output, such as the risk of compressor surge and limitations on the gas turbine torque and turbine inlet temperatures.

A higher mass flowrate through the turbine stage will increase the pressure at the turbine inlet and thus also at the compressor outlet, so that the compressor can run into surge, with the air flow no longer maintained. The amount of pressure increase the compressor can tolerate is defined as the compressor surge margin, which depends on the design of a given compressor. There are several other possible strategies to resolve the surge limitation problem:

- Modify the gas turbine of the GT: The turbine itself can be modified to have an increased cross-sectional area to allow a higher flowrate.
- Modify the compressor of the GT: With an additional compressor stage the gas turbine can operate at a higher overall pressure ratio without surge problems.
- Bleeding off air from the compressor: This solution is possible for plants with air integration. Bleeding off more air mass flow than the mass flow of the nitrogen brought back from the ASU to the turbine reduces the mass flow through the turbine. Air integration therefore provides the potential to use gas turbines which would otherwise need to be redesigned to work with syngas (Maurstad 2005).

#### 7.6.6.2 Air Separation Unit (ASU)

The oxygen supply to a gasifier is one of the most expensive single parts of any gasification process. The capital cost of the ASU, with its associated compressors, is about 10–15% of the total plant cost. The commercial technology used for oxygen production in IGCC plants is cryogenic air separation, which may be defined as the separation of air into component gases by distillation at low temperatures. Cryogenic air separation has single-train  $O_2$  production capacities of 3,200 t/day and is recognised for its high reliability.

The major energy requirement of the process is for the air compression, which is in the order of magnitude of 5–7% of the gross generator output. Typically, the air to the ASU is compressed to around 5 bar, and the oxygen (typically 95%  $O_2$ , 3.5% Ar and 1.5%  $N_2$  by volume) and nitrogen product streams become available at around 1 bar. The process may also operate at elevated pressure so that part or all of the ASU air is supplied from the gas turbine compressor. In this case, the ASU product streams are at around 5 bar, so this reduces the recompression work.

Alternative processes for air separation are limited in capacity and achievable oxygen purity. Pressure swing absorption units are available up to a capacity of about 140 t/day, but they can only reach a purity of about 95%. The product quality obtainable with polymer membrane technology is about 40% oxygen, with capacities of up to 20 t/day. Oxygen purity is a limiting factor for chemical applications. Most probable gasification applications for polymer membrane technology and pressure swing absorption are biomass power applications, where the sizes are also at the lower end of the scale. Both technologies have the advantage of a quick start-up compared to cryogenic units (Higman and van der Burgt 2008; Maurstad 2005).

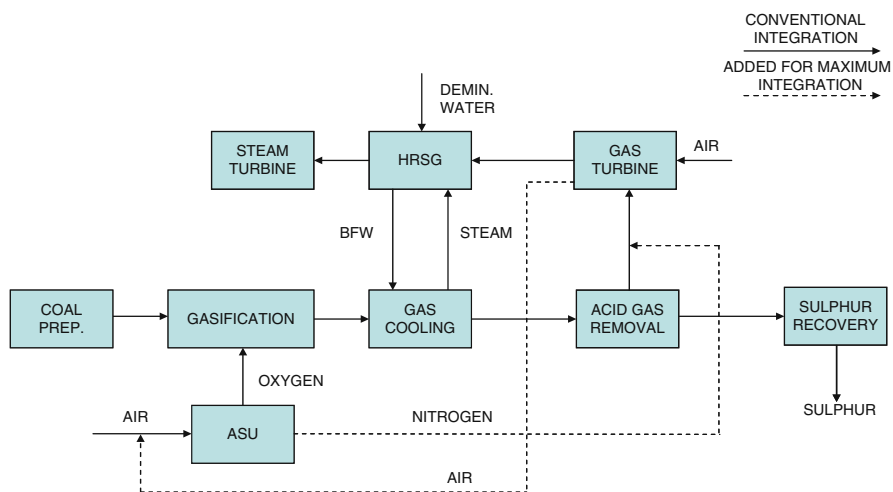
### 7.6.6.3 Integration

In a natural gas fired gas and steam turbine power plant, the gas turbine and steam processes are linked to each other only by the flue gas feed to the heat recovery steam generator. In contrast, in an IGCC power plant, there are several possibilities for coupling the air separation unit, the gasifier, the gas turbine and the steam generator. The purpose of integration – hence the term “integrated gasification process” – is to maximise the efficiency. A high degree of integration, however, can imply disadvantages affecting the operating performance at different loads and diminish the availability of the process as a whole.

Figure 7.83 shows the different possibilities for coupling:

**Steamside integration:** In an IGCC power plant, steam is generated in the heat recovery steam generator (HRSG), the gasifier and the raw gas cooler. In a natural gas fired HRSG, the flue gas temperature determines the live steam temperature via the pinch point of the heat exchanger. This problem is diminished in an IGCC plant by the large evaporative surfaces of the syngas cooler, which can superheat more saturated steam in the waste heat boiler. By doing so, the efficiency of the steam production rises from about 40 to 42%. Steamside integration is applied in all demonstration plants.

**Airside integration:** About 15–25% of the total air flow has to be fed to the air separation unit (ASU) for the generation of oxygen; the rest serves to oxidise the syngas in the gas turbine combustor. It is deemed full integration if the total air flow needed for the ASU is drawn off after the gas turbine compressor (GT compressor). The better compression efficiency of the gas turbine helps to reduce the energy demand for the compression as a whole. The air separation unit is then operated under pressure; the pressure rise of the oxygen up to gasification pressure is



**Fig. 7.83** Integrated IGCC power Plants – level of integration (from Higman and van der Burgt 2008, © 2008, with permission from Elsevier)

therefore smaller. The degree of (air) integration is usually defined as the percentage of the total ASU air required coming from the GT compressor. The two existing US IGCC demonstration plants started with 0% integration, while the two European plants had 100% integration.

The possible benefits of integration are an increased efficiency, increased power output and reduced capital cost (e.g. savings on the ASU air compressor). The drawbacks of high integration are a possible reduced availability of the process, less operational flexibility, lengthy start-up times and the fact that the ASU cannot start without the GT running. An integration of 100% will always yield the maximum efficiency, but not necessarily the maximum power.

Because of the lower process availability and operational flexibility during start-up and shutdown, a partial integration scheme is implemented in newer plants as a compromise between availability and operational flexibility on the one hand and efficiency on the other. In such schemes, only part of the air flow for the ASU is taken from the gas turbine, and the rest of the air is fed by way of a separate compressor.

*Integration on the nitrogen side:* In order to reduce the flame temperatures in the gas turbine and the nitrogen oxide emissions, nitrogen is fed before or into the gas turbine combustor. Similarly to air integration, prior pressurisation of the nitrogen reduces the necessary pressure increase up to the gasification pressure. Even if the air integration is 0%, it may still be beneficial to use nitrogen from the ASU for NO<sub>x</sub> reduction (Geosits and Schmoe 2005; Higman and van der Burgt 2008; Eurlings and Ploeg 1999; Maurstad 2005).

## ***7.6.7 State of the Art and Perspectives***

### **7.6.7.1 IGCC Plants in Operation**

The worldwide installed gasification capacity of about 70 GW<sub>th</sub> predominantly serves to make chemical products and fuels; only about 20% of the capacity is used to generate electrical power. For power generation, residues from refinery processes are mostly used – the generation of electrical power by coal in IGCC power plants worldwide is limited to a few plants with a total fuel capacity of 3 GW (NETL 2007; Minchener 2005). IGCC technology for coal was demonstrated, using public funding, at several locations in the 1990s in Europe and the USA. The following plants were part of those demonstrations and are being operated commercially at the moment:

- In 1994, the 253 MW<sub>el</sub> (semi-)commercial IGCC power plant Demkolec was commissioned at Buggenum, the Netherlands. The net generating efficiency of this power plant, which uses Shell gasification technology, is 43.2%.
- In 1995, the 252 MW<sub>el</sub> Wabash River IGCC went into operation in Terre Haute, Indiana (USA), based on the E-Gasification process. Wabash River was a repowering of a small steam power plant and not a greenfield project. The IGCC has a net efficiency of 39.9%.



- In 1996, a 250 MW<sub>el</sub> IGCC was commissioned in Polk County, Florida (USA). This IGCC power plant, based on Texaco gasification technology, has a net generating efficiency of 38.0% (Tampa.Electric 2002; Tampa.Electric 2004).
- In 1996, a 318 MW<sub>el</sub> IGCC went into service at Puertollano (Spain). Until 1998, the power plant was operated using natural gas as a (secondary) fuel (Hannemann et al. 2003; Mendez-Vigo et al. 1998; Cortes 1999). When the gasifier was ready for operation in 1998, hard coal and petroleum cokes (50/50%) became the main fuels. Puertollano makes use of Prenflo gasification technology and has a net efficiency of 45%.

The plant data is compiled in Table 7.18. The two European plants, due to their gas turbines, dry feeding, dry quenching and high degree of integration of the air separation unit, have a higher efficiency.

### 7.6.7.2 Description of the Puertollano Plant

The IGCC plant in Puertollano is the biggest coal-based IGCC power plant in the world, having a net capacity of 305 MW<sub>el</sub> and a design efficiency of 45% at a condenser pressure of 0.0715 bar. The fuel used is a mixture, consisting half of a Spanish coal with a very high ash content (47% ash) and half of a high-sulphur petrol coke. The efficiency is highly dependent on the fuel; running with the petrol coke alone, an efficiency of only 42% is achieved. Despite the high-sulphur fuel, the emissions of SO<sub>x</sub> remains below 20 mg/Nm<sup>3</sup> (at 6% O<sub>2</sub>), which is below the EU limit by one order of magnitude.

The entrained-flow gasifier is fed with oxygen of 85% purity, and at high temperatures (above 1,600°C) the finely milled fuel gets converted. The fuel is blown in pneumatically with a nitrogen stream which is tapped after the air separation unit. The pressure in the reactor is 25 bar; the temperature at the outlet of the gasifier is about 1,600°C. The employed gasification system is a Prenflo entrained-flow gasifier, which is roughly similar in construction to the Shell gasifier. About 30–50% of the cold syngas is recycled to quench the hot syngas from the gasifier to about 900°C. A convective heat exchanger further reduces the temperature to about 200°C. The particles are separated from the cooled raw gas in a ceramic cartridge filter, and there is the potential to return the fly ash to the gasifier by way of a lock-hopper system, thus removing almost the entire ash load from the process as unleachable slag. Subsequently, the raw gas is subjected to wet gas cleaning, consisting of an MDEA scrubber, an upstream COS hydrolysis unit and a Claus tail-gas cleaning process for sulphur production. The residual sulphur content of the coal gas is then less than 25 mg/m<sup>3</sup>. After the cleaning process, the syngas is diluted with nitrogen from the air separation unit and burned in the gas turbine combustor. The flue gases, with a gas turbine entry temperature of 1,150°C, expand in the gas turbine. The remaining heat of 500°C as it exits the turbine is used in the HRSG (Coca 2003).

The air separation unit has a high degree of integration and is fed with a partial flow from the air compressors. Part of the nitrogen stream produced during air

**Table 7.18** Data for IGCC power plants in operation (Hannemann et al. 2003; Lako 2004; Tampa\_Electric 2002; Tampa\_Electric 2004; Holt 2003; Coca 2003)

IGCC power plant	Wabash river	Buggenum	Puertollano	Tampa
Year (operational)	1995	1994	1998	1996
State/Country	Indiana/USA	The Netherlands	Spain	Florida/USA
Feedstock	Hard coal	Hard coal	Lignite and petr. coke	Hard coal
Secondary fuel	–	Natural gas	Natural gas	
Gasification process	E-Gas	Shell	Prenflo	GE
Level of integration	Low	High	High	Low
Raw gas (after the gasifier)				
Fuel gas temperature [°C]	300	300	302	330
H <sub>2</sub> [%vol.]	34.4	25.5	22.1	36.4
CO [%vol.]	45.3	62.7	60.5	42.8
CO <sub>2</sub> [%vol.]	15.8	2.2	3.9	14.4
N <sub>2</sub> [%vol.]	2.9	8.8	12.5	3.3
Ar [%vol.]	0.6	0.8	1.0	0.9
H <sub>2</sub> /CO ratio (vol.)	0.76	0.4	0.36	0.85
HHV [MJ/m <sup>3</sup> ]	10.3	10.4	10.5	9.9
Fuel gas (fired in the gas turbine)				
N <sub>2</sub> [%vol.]		42.0	53.1	
H <sub>2</sub> O [%vol.]		19.1	4.2	
LHV [MJ/m <sup>3</sup> ]		4.3	4.3	
Emissions				
SO <sub>2</sub> emission [mg/m <sup>3</sup> ]	40	35	25	40
NO <sub>x</sub> emission [mg/m <sup>3</sup> ]	100	25	150	100
Integration				
Air extracted from GT related to ASU [%]	0	100	100	0
Related to compressor [%]	0	16	18	0
Nitrogen integration [%]	0	100	100	100
Output				
Gas turbine [MW]	192	156	179	192
Steam turbine [MW]	96	128	137	120–135
Net power output [MW]	252	253	276	250
Net efficiency [LHV]				
High-quality hard coal [%]	39.9	43.2	45.0	38.0
Lignite and petcoke [%]	–	–	42.0	
Petroleum cokes [%]		–	–	
Natural gas [%]	–	52	52.4	

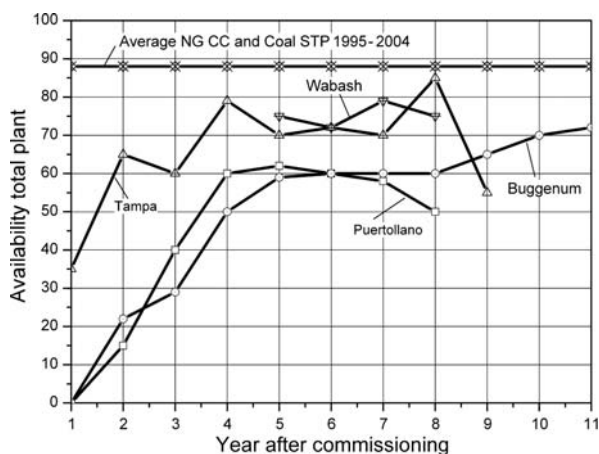
separation is used to transport the dry and milled fuel mixture into the gasifier, while another part of it cleans the raw gas dedusting cartridge filters. The remaining nitrogen stream is mixed in with the cleaned coal gas before entering the gas turbine combustor to lower the combustion temperature and comply with the NO<sub>x</sub> emission limit without flue gas cleaning.

In Fig. 7.86, an optimised IGCC configuration based on the configuration of the plant in Puertollano is shown.

### 7.6.7.3 Process Availability and Costs of IGCC Plants

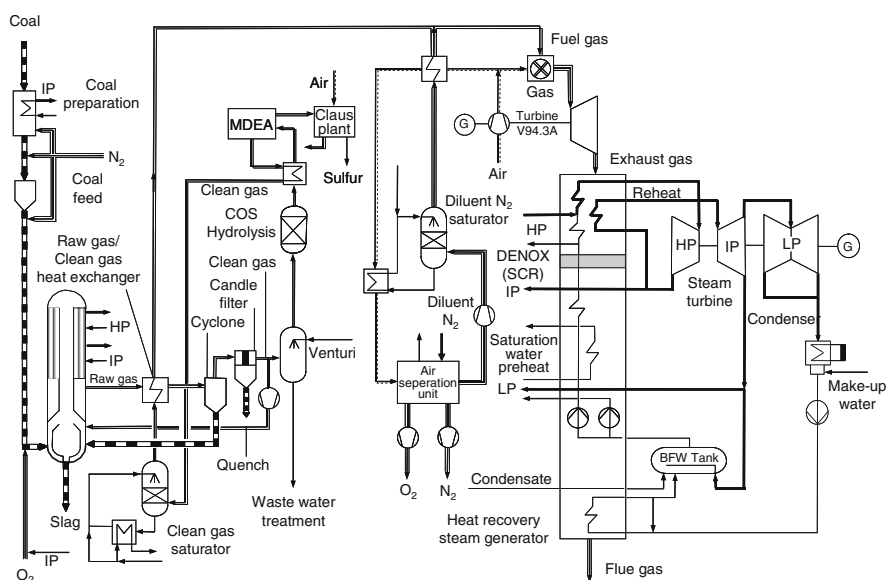
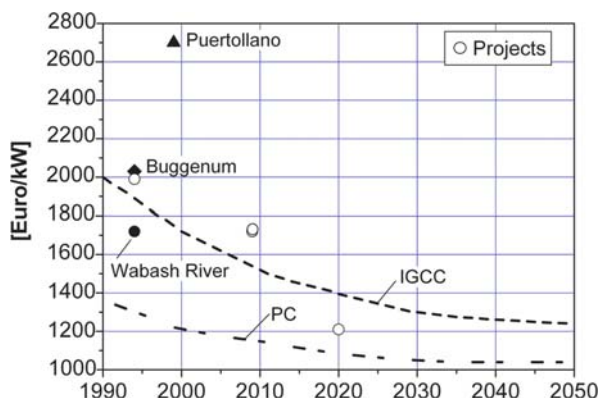
In all the IGCC power plants in operation, a great number of problems arose in the first years of operation in relation to the gasifier, the gas turbine and the gas cleaning. The high integration degree of the ASU, too, gave rise to difficulties and resulted in the use of new control techniques. Today, the process availabilities of the power plants range between about 70 and 80% and are thus markedly lower than the availabilities of steam power plants. Figure 7.84 shows this graphically, making it clear that the main problem of this technology is its lower availability. The ability to compete with other power plant technologies requires availabilities higher than 90%. IGCC plants in refineries show higher availabilities when tried-and-tested technologies and lower degrees of integration are used (Folke 2006; Higman 2005). New data from Buggenum with availabilities of 90% underlines that from an engineering point of view it should be possible to achieve similar availabilities than in conventional power plants.

Another problem is the considerably higher cost of IGCC power plants. Figure 7.85 pits the capital costs of the installed IGCC power plants against the capital costs of conventional pulverised coal fired steam power plants, using Euros in 2004 as a basis. The capital costs of the erected plants were between 30 and 100% above the costs of conventional steam power plants; currently, it is assumed that costs are 40% higher. The expectation is, however, that this cost difference will decrease as advancement along the learning curve for IGCC technology occurs (Lako 2004). In addition, if CO<sub>2</sub> capture is considered, it is expected that IGCC power plants with CO<sub>2</sub> capture and steam power plants with CO<sub>2</sub> capture will have roughly the same level of capital costs.



**Fig. 7.84** Process availability of existing IGCC plants (Folke 2006)

**Fig. 7.85** Cost of IGCC plants in comparison to conventional steam power plants (Lako 2004)



**Fig. 7.86** Process flow diagram of IGCC 98 (Pruscek 2002)

#### 7.6.7.4 Efficiency Potential

IGCC power plants in operation today achieve electrical efficiencies of up to about 45%. Using today's state-of-the-art technology, without CO<sub>2</sub> removal, IGCC power plants could be designed and built with an electrical efficiency of over 50%. If the classical single components and the overall IGCC concept were to be further optimised, electrical efficiencies of 55% could be achieved within 15 years. Estimates of efficiency are based both on achieved efficiencies at operating plants (and the potential to optimise them) and on the expected future advancements in technology, particularly for the gas turbine.

Extensive studies on the potential of IGCC power plants were carried out by Pruscek, who, within the framework of European projects, and comparing to the IGCC plant in Puertollano, investigated the design and the efficiency of an IGCC power plant feasible in 1998 (Pruscek 1998). Its configuration, shown in Fig. 7.86, is similar to the configuration of the Puertollano IGCC power plant. The entrained-flow gasification is operated at a pressure of 29 bar with 95% oxygen as the oxidiser. The gas exiting the gasifier first gets cooled to about 900°C with recirculated raw gas, then the remaining flue gas heat is used to generate steam and to reheat the cleaned fuel gas. The air separation unit is fully integrated, so that the fresh air is also compressed in the gas turbine compressor and part of the waste nitrogen is mixed back into the fuel gas before entering the gas turbine combustor. The waste nitrogen and the cleaned fuel gas are humidified with the necessary water fraction by saturators before the mixing is performed, at a temperature as low as possible. At the given ambient conditions and at a gas turbine entry temperature of 1,250°C (ISO), the IGCC achieves an efficiency of 51.5%. The efficiency owes its increase, in essence, to the raising of the gas turbine entry temperature to 1,250°C, as well as further measures such as the increase of the steam conditions and more intensive fuel gas preheating (Haupt et al. 1998; Pruscek 1998; Pruscek et al. 1997).

Over and above the 1998 IGCC concept design, there are considerable potentials for increasing the efficiency of IGCC power plants. Applying the measures shown in Fig. 7.87, efficiencies up to 58% could be achieved. It becomes clear that development of gas turbines would yield the largest efficiency increases. Hot dry gas cleaning would raise the efficiency by about 0.7% (Pruscek 1998).

### 7.6.7.5 IGCC Concept Designs with CO<sub>2</sub> Removal

IGCC power plants can separate CO<sub>2</sub> with few additional components and can be regarded as the most advanced power plant technology for CO<sub>2</sub> separation, because the necessary additional components (CO shift and CO<sub>2</sub> scrubbing units)

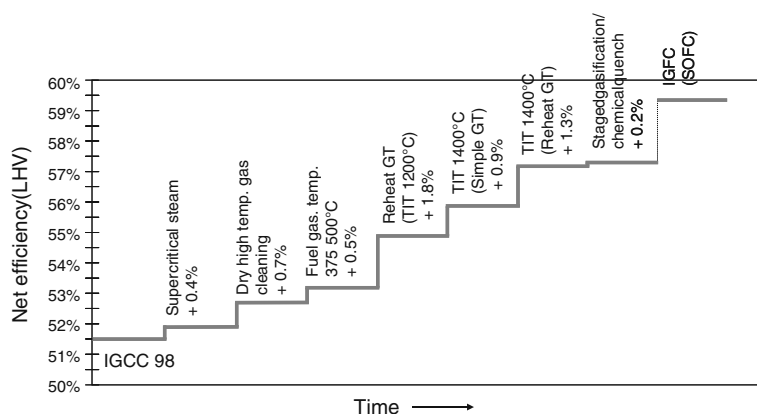


Fig. 7.87 Potential future development of IGCC power plants (Pruscek 1998)

are employed already for the production of hydrogen. An IGCC design with CO<sub>2</sub> capture is shown in Fig. 8.22. Expectations are that the efficiency will drop by about 8–10% compared to a process without CO<sub>2</sub> capture – starting out from the IGCC 98 concept, having 52% efficiency, the efficiency decreases to 42%. In Chap. 8, the IGCC technology with CO<sub>2</sub> removal is compared to competing methods for CO<sub>2</sub> separation during power production.

Various projects examining the design and construction of a CO<sub>2</sub>-free power plant using IGCC technology are currently in progress, such as the following:

- RWE: Construction of a 360 MW<sub>el</sub> lignite IGCC-CCS power plant by 2014 (Lambertz and Ewers 2006). The current design results in low efficiencies in the range of about 35% because first priority has been given to the plant availability. For this reason proven technologies and a low level of integration have been chosen.
- FutureGen (USA): Construction of a 275 MW<sub>el</sub> IGCC-CCS power plant, a project lasting until 2012 (FutureGen 2007)
- GreenGen (China): Development of its own coal gasification technology by 2009 followed by construction of a 400 MW<sub>el</sub> demonstration plant for hydrogen production with CO<sub>2</sub> capture (Folke 2006).

#### 7.6.7.6 Long-Term Perspectives

Further development must aim to eliminate the disadvantages of IGCC power plants, namely the low availability and the high costs. If these problems can be solved, IGCC could provide a substantial increase in the efficiency of power generation from coal.

In gasification, the major part of the fuel energy gets converted into chemically bound energy and a smaller part into heat. A level of conversion into chemically bound energy as high as possible and a heat exploitation as efficient as possible should be aimed at.

The quality of the conversion into chemically bound energy is described by the cold gas efficiency. The maximum conversion of fuel energy into chemical energy can be achieved using chemical quenches or by internal heat utilisation. Gasification methods should be sought that combine the benefit of fluidised bed gasification (high cold gas efficiency) with the benefit of entrained-flow gasification (ash removal, fuel flexibility, compactness).

The utilisation of the heat of hot syngases would be optimal if the gases could be fed directly into the gas turbine. However, for gas cleaning, these gases have to be cooled down to very low temperatures. The heat removed when cooling to these temperatures can be partly used, being converted into mechanical energy with the lower efficiency of the steam cycle. On top of this, the requirements of gas cleaning involve cooling and heating processes that cause additional exergy losses. In order to avoid them, gas cleaning processes need to be developed that run at higher temperatures and in dry conditions to the greatest possible extent. The aim

is not to cool the gas below the temperature of the following step in the process, so that a continuous extraction of useful heat is possible.

A further significant increase in the energy efficiency can be expected by using hydrogen membranes. With  $H_2$  membranes, only a stoichiometric steam-to-CO ratio would be necessary. This could decrease the demand of steam significantly, while also making it possible to avoid the cooling to the thermodynamically necessary low temperatures for the shift reaction, thus providing a hotter gas for the burner of the gas turbine. In this respect, high-temperature membrane shift reactors are desired, which, in one unit, perform both the gas conversion (shift) and the gas separation.

The long-term development objective for a  $CO_2$ -free IGCC power plant with the highest possible efficiency is therefore a high-pressure, high-temperature gasifier with integrated hot gas cleaning,  $H_2$  separation by catalytic high-temperature membranes at the highest possible temperatures and an  $H_2$  gas turbine being fed with hot fuel gas.

In the long term, it will also be reasonable to combine gasification and solid oxide fuel cells (SOFCs). The technology suited in particular for this process is allothermal fluidised bed gasification, which utilises the waste heat of an SOFC to supply gasification heat.

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